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Targeted Gas Spectroscopy Using Tunable VCSELs

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June 15, 2011

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Oakridge, TN, United States
June 27, 2011 through June 30, 2011

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Targeted Gas Spectroscopy Using Tunable VCSELs

JOWOG 28 Main Meeting
Y -12 National Security Complex, Oak Ridge
6/27-30/2011



Tiziana Bond, Mihail Bora, Allan Chang, Jim McCarrick

Lawrence Livermore National Laboratory, P. O. Box 808, Livermore, CA 94551

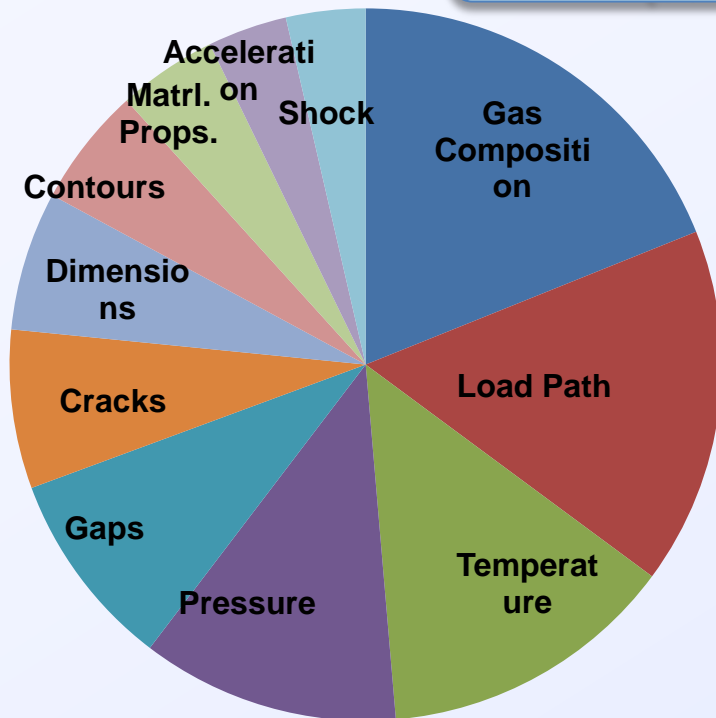
This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. LLNL-CONF-487792

- **Motivation**
- **Background**
- **Measurements (Setup and results)**
 - Basic Absorbance
 - Wavelength Modulation
- **Conclusion**
 - Steps forward



Persistent Surveillance of complex systems by embedded sensing: a new paradigm of aging awareness and monitoring

Gas sensing will provide early detection of a broad range problems (decomposing components, corrosion, failures, etc)



Gas Sensing System Requirements

- Detection of mixtures of unknown species
- High sensitivity - ppm level of detection
- High selectivity – broad range of gas molecules (e.g. diatomics, organics, inorganics)
- Fiber optic compatible
- Rugged, robust system (lifespan = decades)
- **Minimize SWaP (size, weight, and power)**

Develop a chip scale sensor spectrometer for minimally invasive monitoring of targeted trace gases

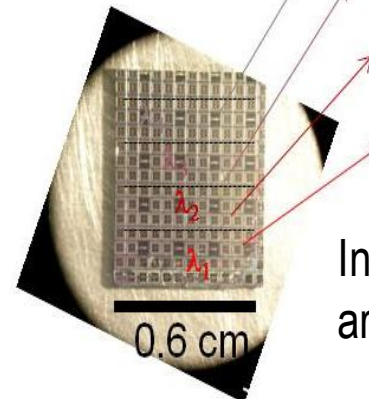
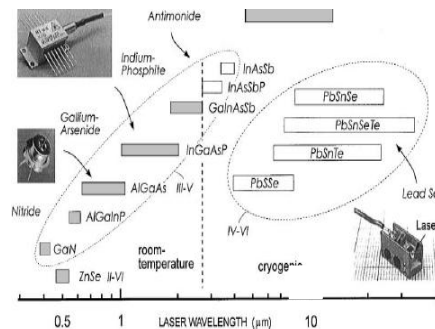
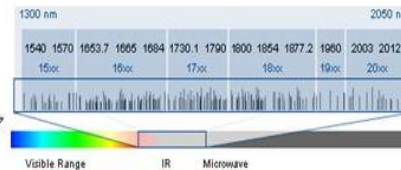
FTIR spectrometer



Vertical cavity
surface emitting laser



1 cm



0.6 cm

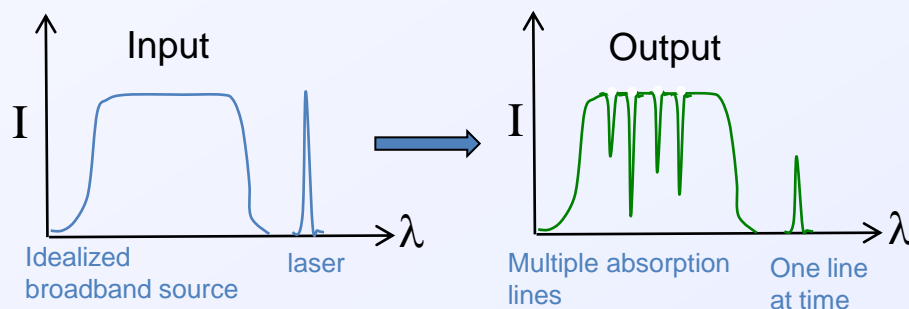
Integrated
array

Approach: Exploit maturity and developments in tunable **VCSEL** technology for broadband **TDLAS** (*Tunable Diode Laser Absorption Spectroscopy*)

Application: atmospheric chemistry, combustion research, space exploration, industrial processing and emission monitoring, and toxic gas detection

VCSEL-based Gas Sensing of Targeted Gases: General Concept

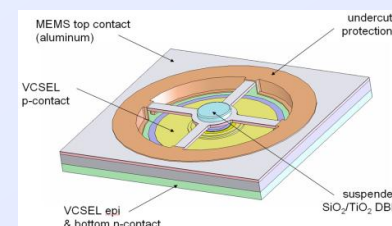
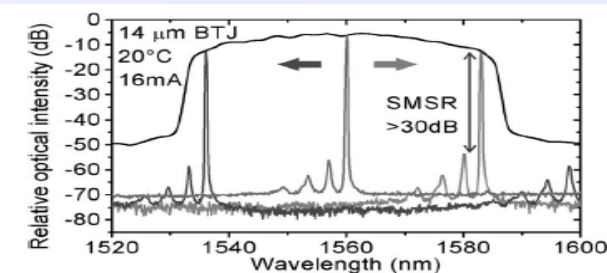
- **Absorption Spectroscopy:** measuring the concentration of gases by the strength of their infrared (IR) absorption fingerprint:



Molecules	λ (nm)
H ₂ O	1390,1802,1854,1870,2360
CO ₂	789,1960,2003,2012
CO	1570, 2360
NO	1800, 2650
NO ₂	680
N ₂ O	1380,1960,2260
CH ₄	1650, 1684,2360
NH ₃	1500
C ₂ H ₂	1520
O ₂	760

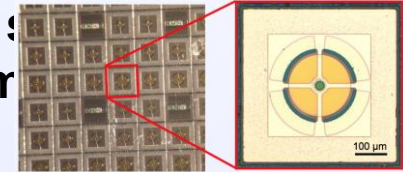
- **Vertical Cavity Surface-Emitting Lasers (VCSELs): compact, low-power**

- Amplified wavelength is selected by cavity properties from a ~50 nm natural range
- Cavity can be tuned thermally (~5 nm max) or via MEMS (10s of nm)
- Bandgap engineering: commercial devices now exist w/ center wavelengths into the mid-IR (2.3 μ) at room temperature



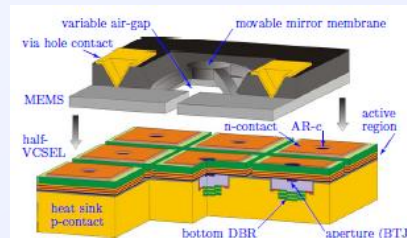
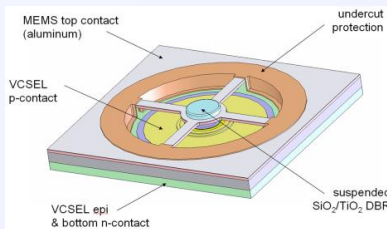
Specific Approach: closed-cavity, open-path VCSELs gas sensing tests

- **Ultimate goal: a battery of tunable devices which cover a** number of targeted gases of interest, combined onto a sm



TDLAS (tunable diode laser absorption spectroscopy)

Collaboration to fabricate open-cavity, MEMS-tunable (R&D) @ 760, 1550, 2300nm



Use of commercial closed-cavity, current-tuned devices w/ external sensing path
COTS @ 760, 1392, 1550, 1854, 2012nm

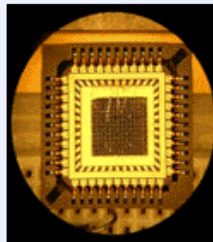
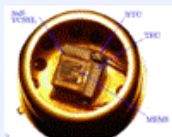


Minimize external path, e.g. via wavelength modulation

LLNL

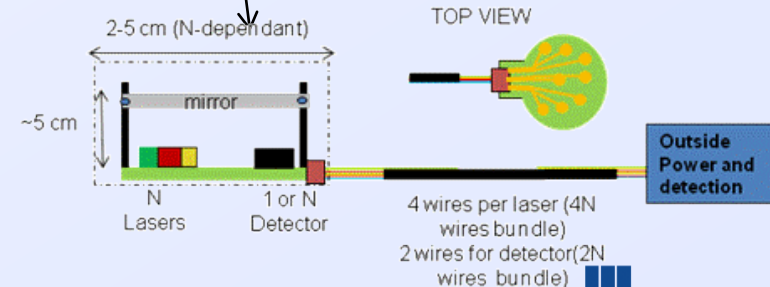
Darmstadt Univ./Schottky Institute

Height ~ 1-2cm
100s devices



1-2 cm

Integration,
Packaging and
Multiplexing



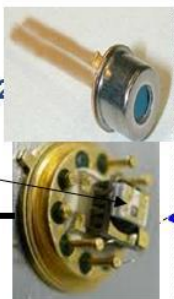
Lawrence Livermore National Laboratory



Closed-cavity, open-path VCSELs gas sensing tests

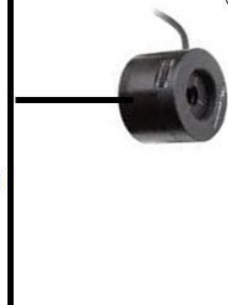
Tunable VCSELs

- ULM-760nm (O_2)
- Vertilas -1392/1854nm/2012 (H₂O and CO₂)
- Thermo-electric cooler



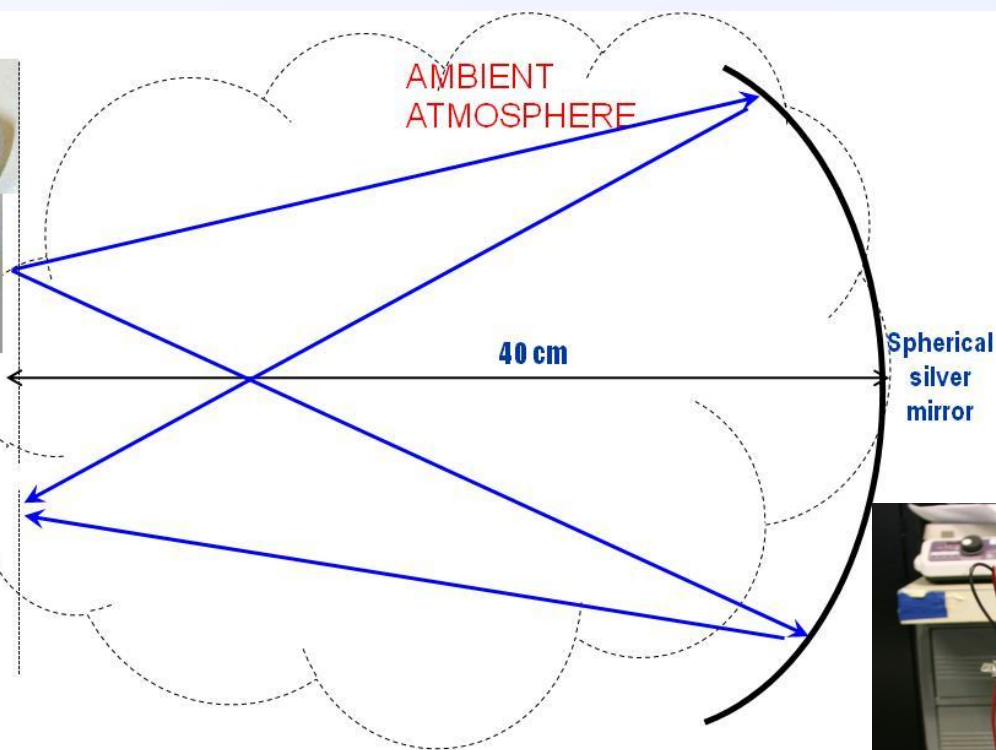
Photodetector

- Newport Si (<1100nm)
- Thorlabs InGaAs (1200-2500nm)

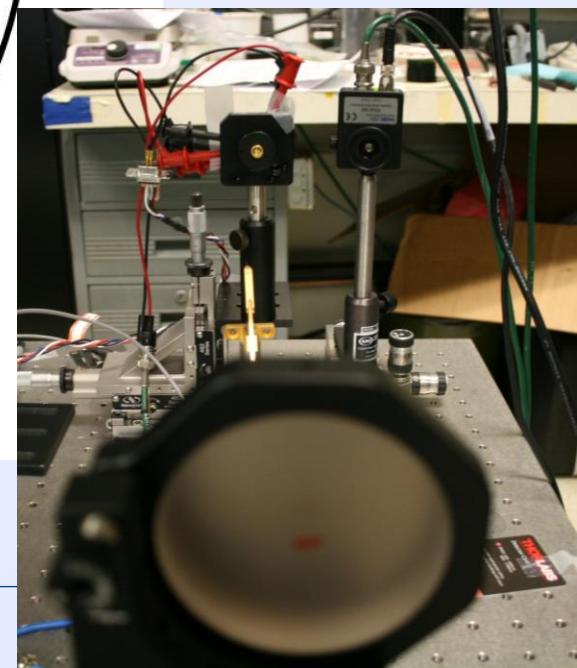
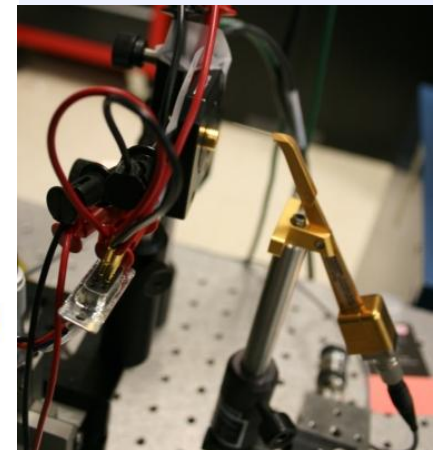


Control Instruments

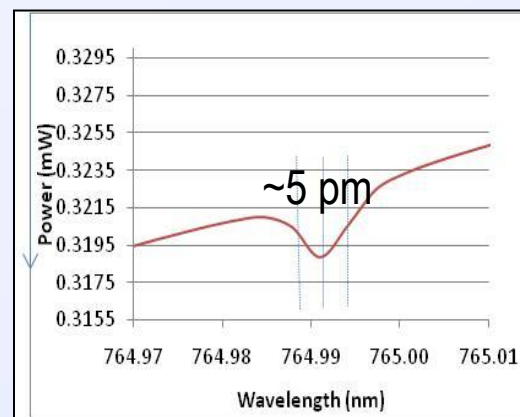
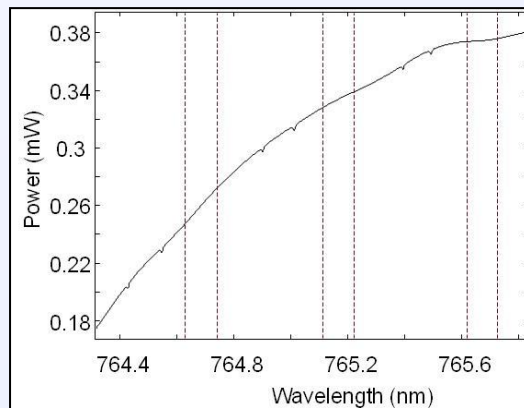
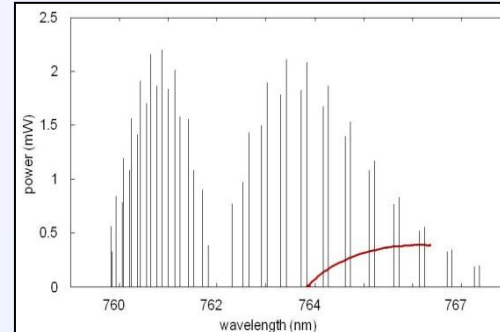
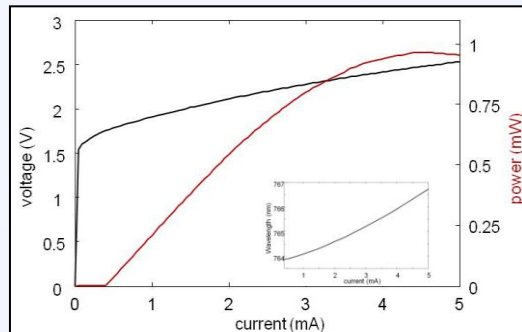
- Keithley Dual Source/Meter (LIV)
- ANDO Optical Spect. Analyzer ((wavelength scan)



Cascade Fiber Probe



O₂ detected in ambient air (21%) with 763nm VCSEL



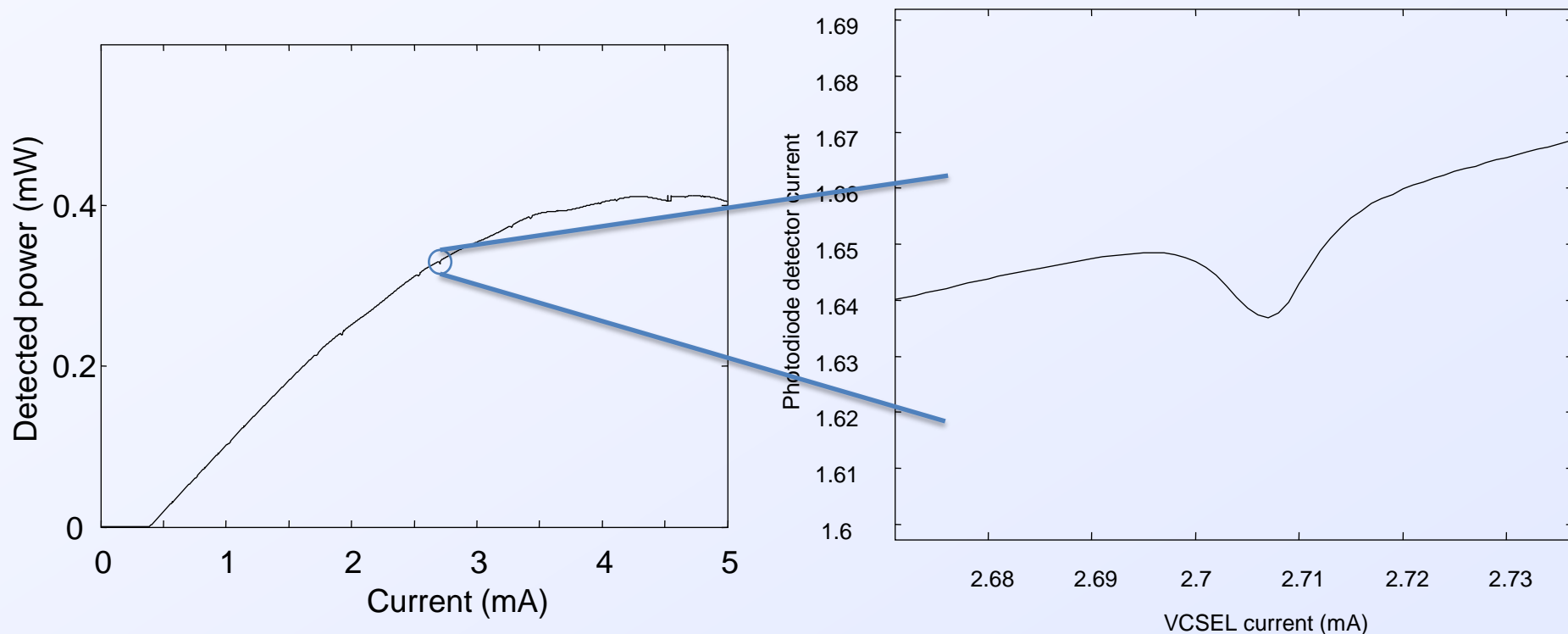
Quench is clearly observable

- Signal change due to oxygen absorbance: $\Delta P/P \sim 1\%$
- Absorption line is above the noise level ($< 0.5 \mu\text{W}$)
- Measurement of O₂ concentrations $> 0.5\%$ are possible
- Scaling to open-cavity possible ($\sim 1 \mu\text{m}$ gap and $Q \sim 10^5$)

$$A = T / T_0 = \exp(-\alpha L_{\text{eff}}) = \exp(-\sigma_g C_g Q L)$$

Preliminary experiments have shown wavelength modulation can greatly improve signal-to-noise

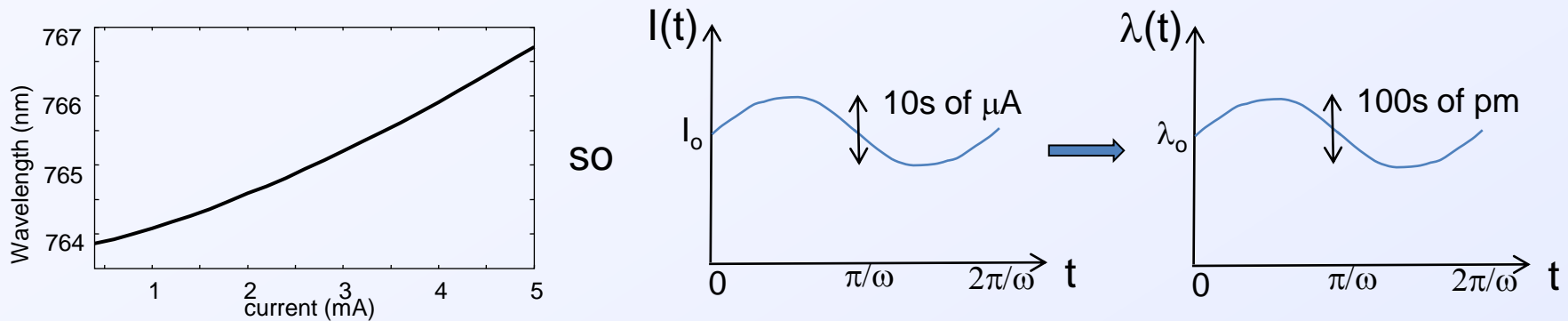
- At low concentrations or with weakly absorbing gases, absorption fingerprint is just a slight bump on the background intensity



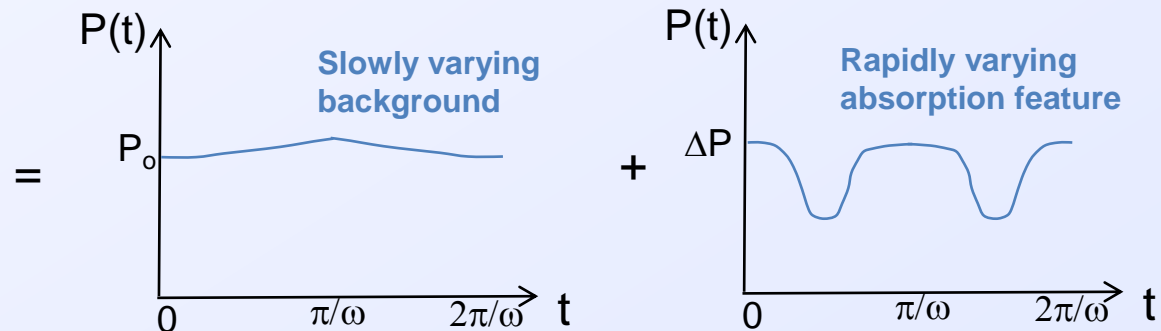
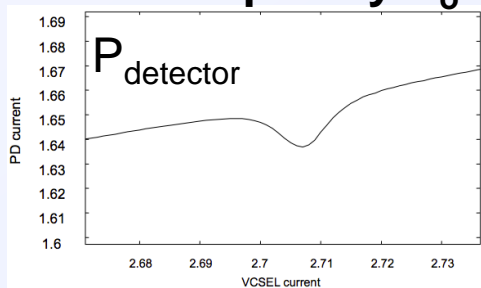
Ambient O₂ detection w/ 763 nm COTS VCSEL

Wavelength modulation pulls the bump out of the background by making it a time-varying signal

- Modulating the laser drive current modulates the output wavelength



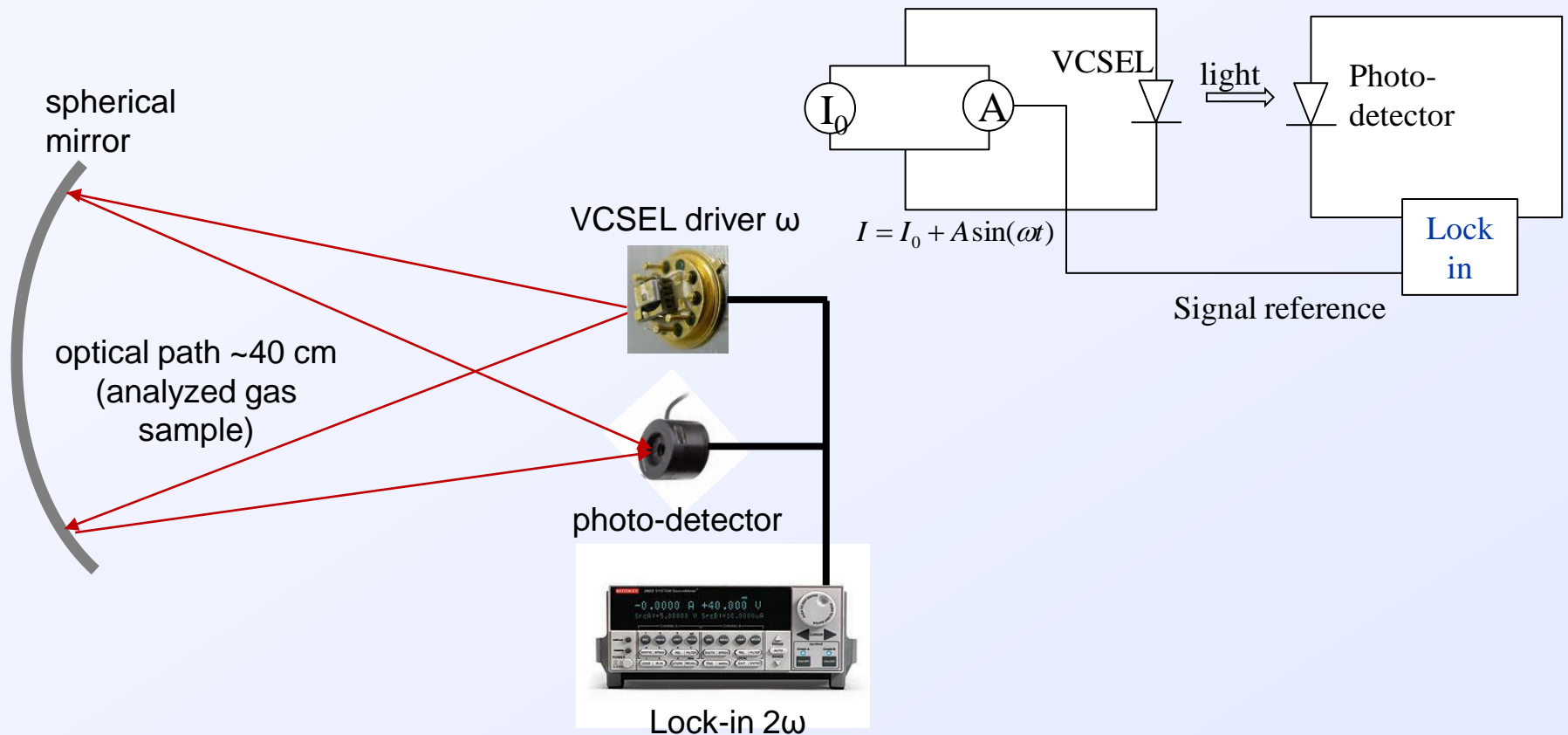
- The detected signal is now periodic in harmonics of the modulation frequency ω_o



- A lock-in amplifier then measures individual harmonic components with high signal-to-noise

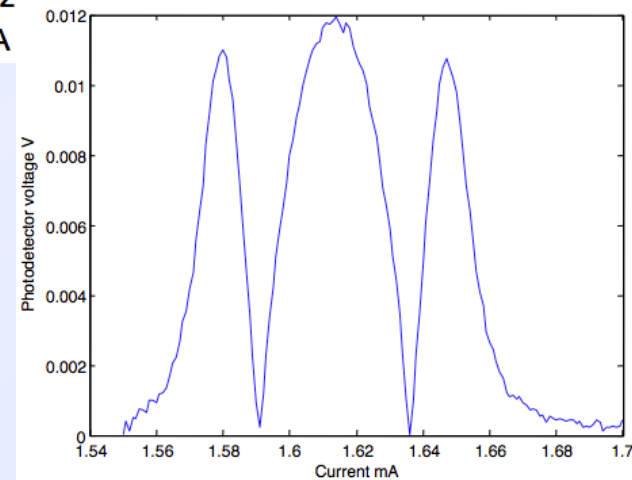
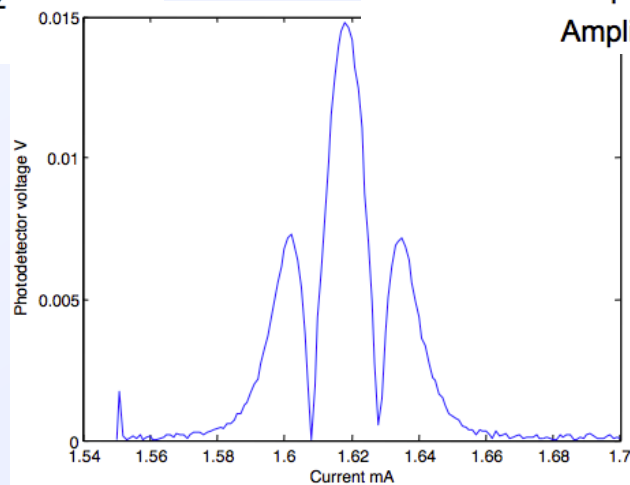
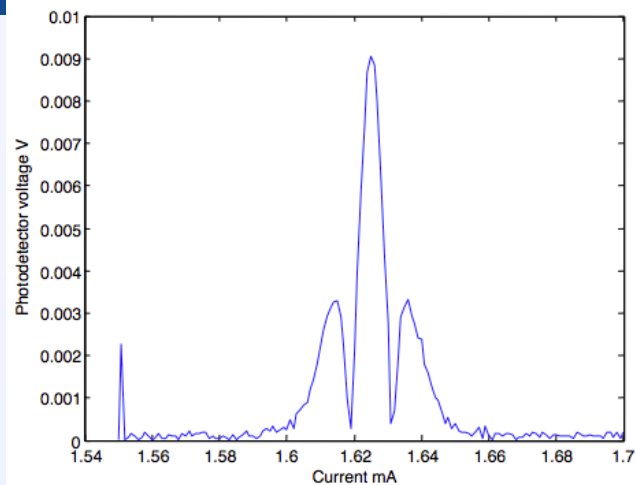
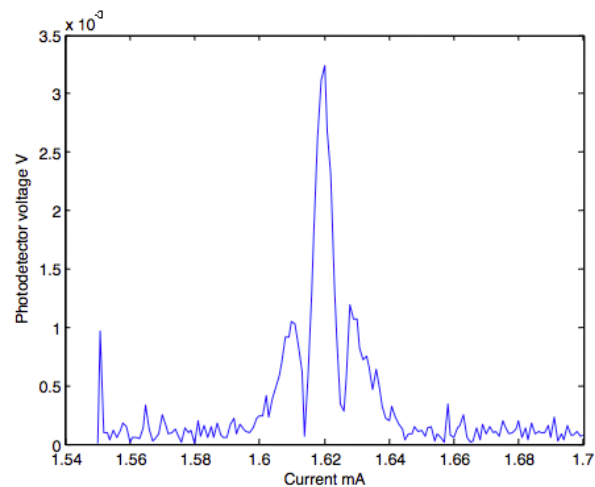
$$P_{\text{lock-in}}(n\omega_o, \phi) = \frac{1}{\Delta T} \int_t^{t+\Delta T} P_{\text{detector}}(t) \sin(n\omega_o t + \phi) dt$$

Modified measurement setup for wavelength modulation spectroscopy (WMS)

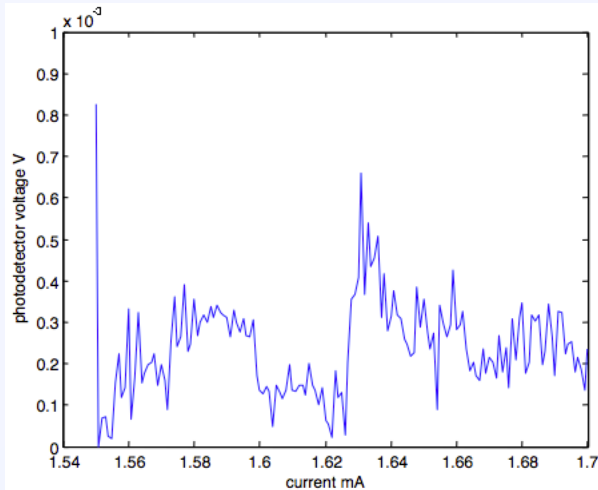


- A series of lock-in measurements have been made at the 2ω component of the photo-detector signal

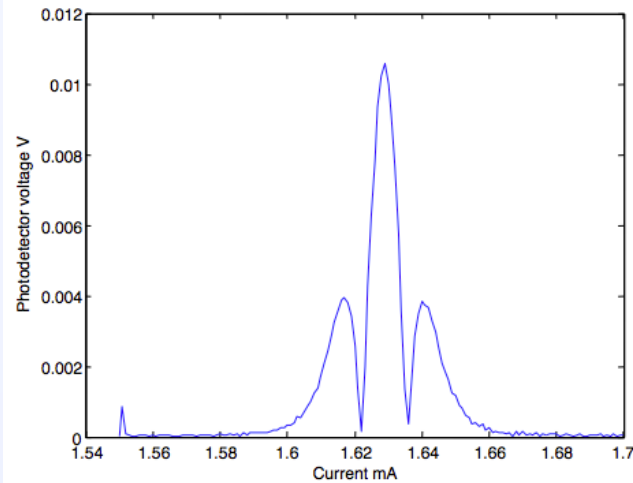
Parameter study in amplitude of current modulation; curves are versus I_0



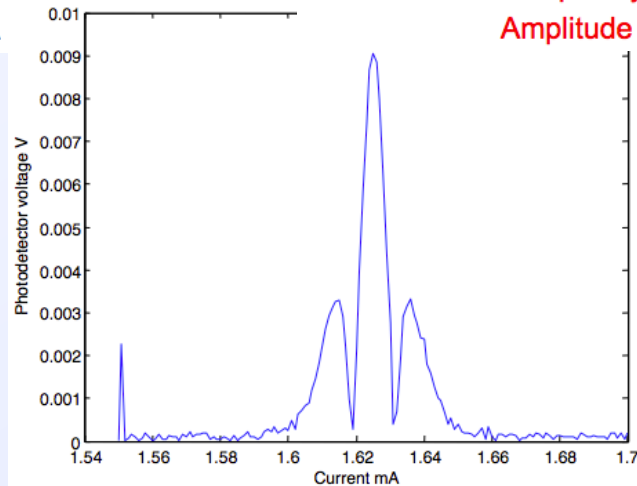
Parameter study in modulation frequency; curves are versus I_0



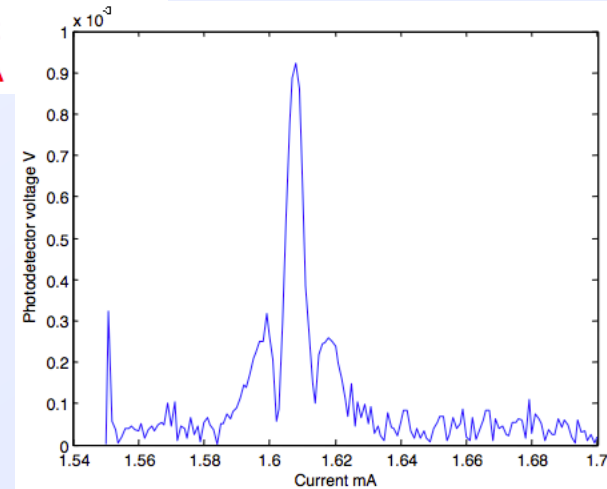
Frequency $\omega=10$ Hz
Amplitude $A = 10 \mu\text{A}$



Frequency $\omega=1$ kHz
Amplitude $A = 10 \mu\text{A}$



Frequency $\omega=10$ kHz
Amplitude $A = 10 \mu\text{A}$



Frequency $\omega=40$ kHz
Amplitude $A = 10 \mu\text{A}$

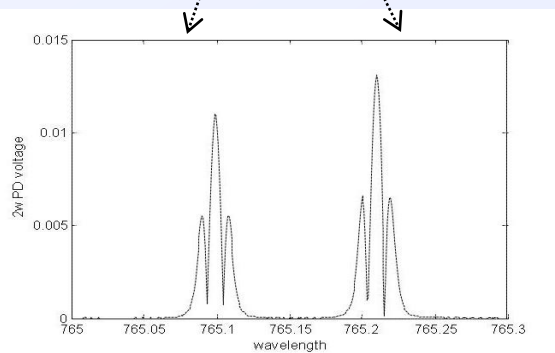
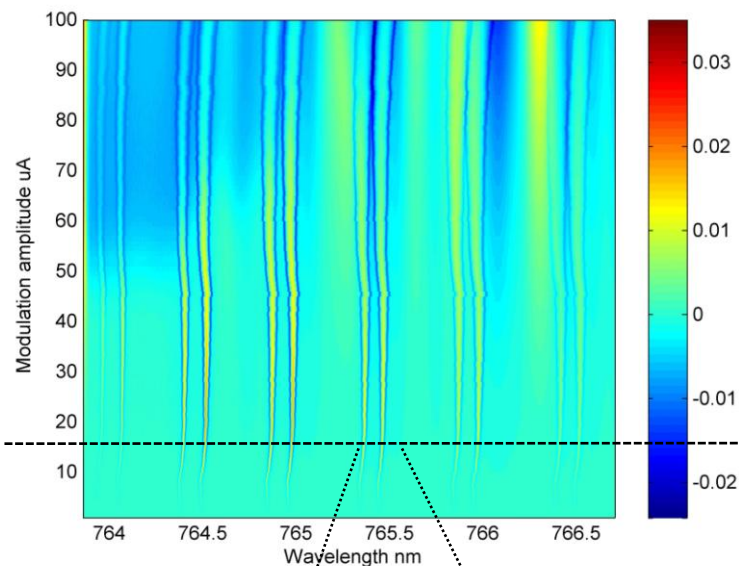
WMS amplitude scan: automated for full range analysis

OXYGEN. Modulation freq 1 kHz; No T control

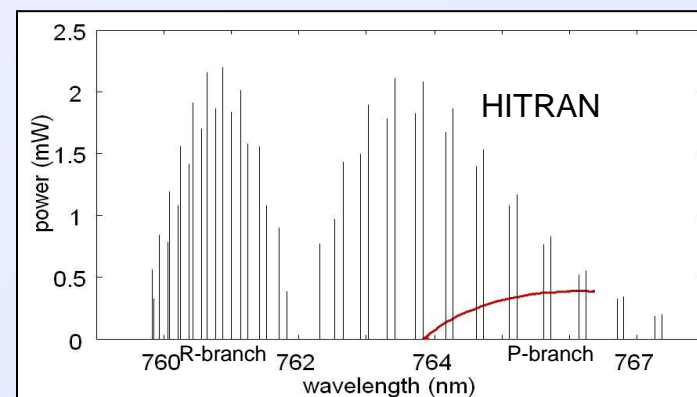
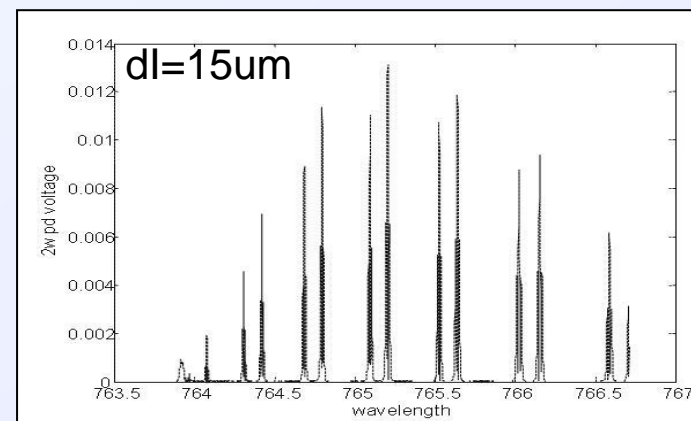
day

night

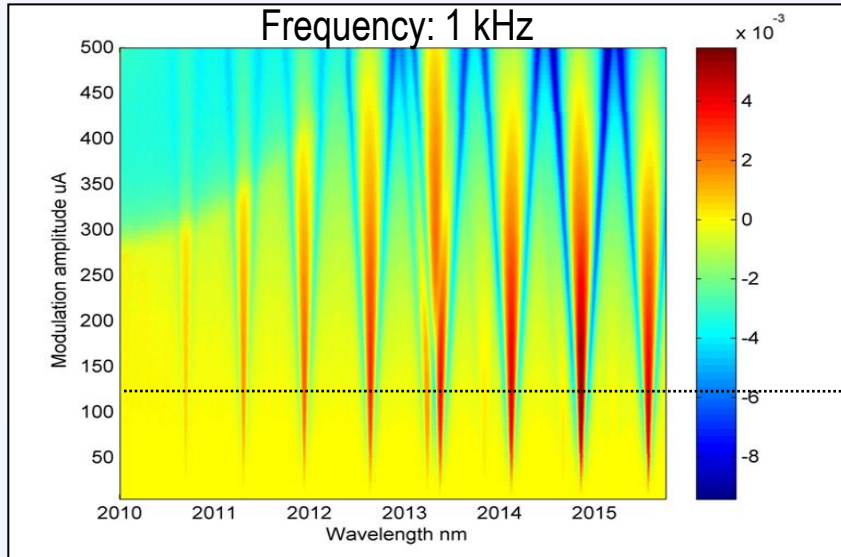
day



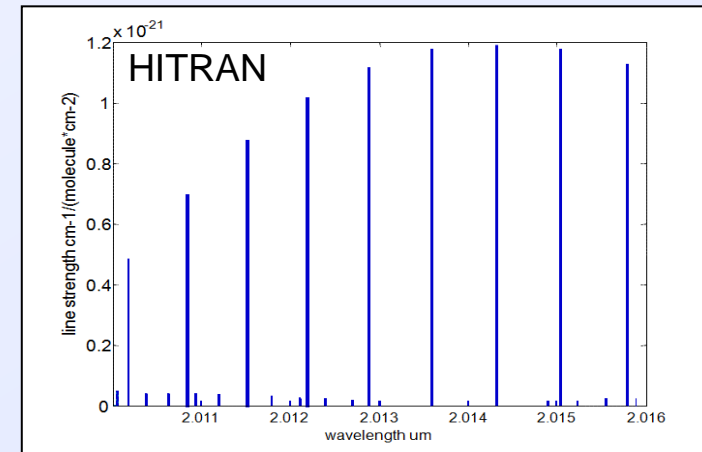
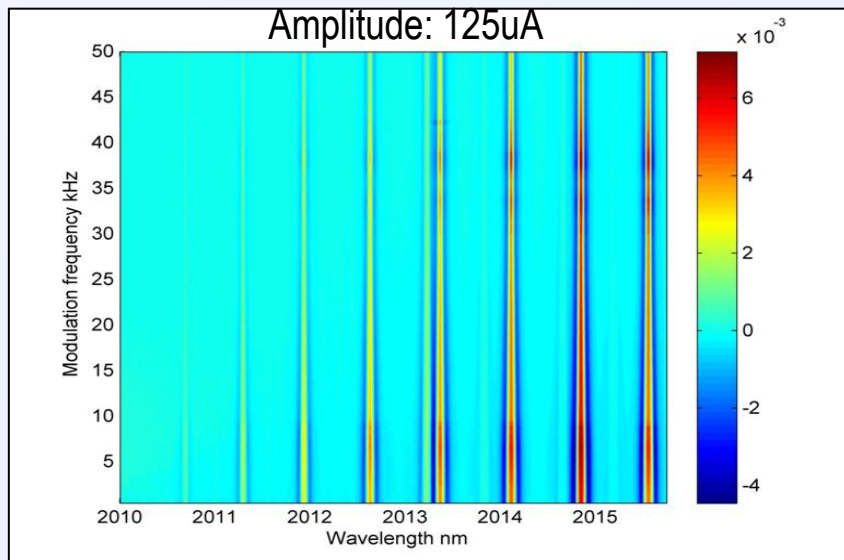
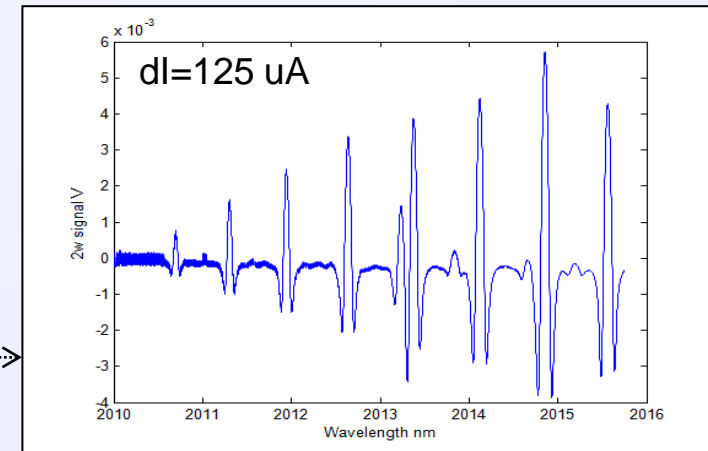
SIGNATURE



WMS of CO₂ in air (~400ppm): amplitude and frequency sweep

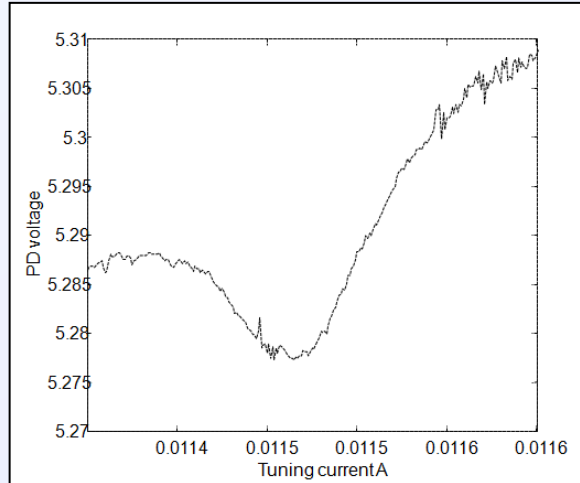
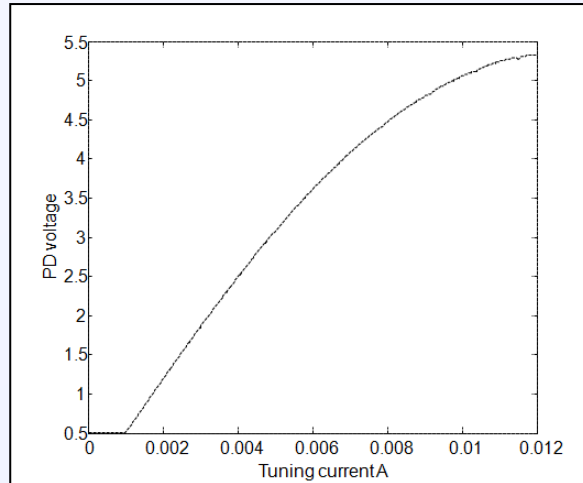


SIGNATURE



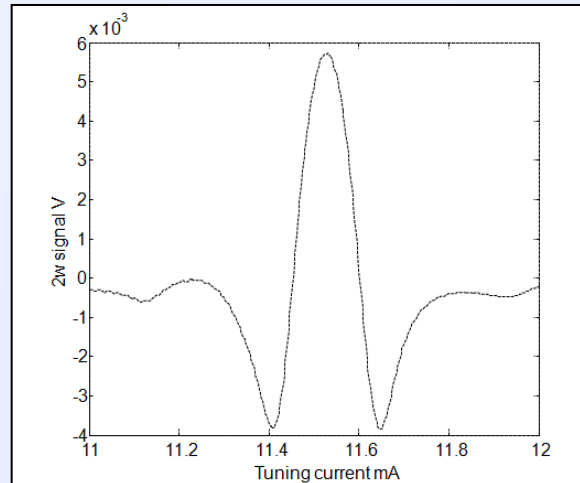
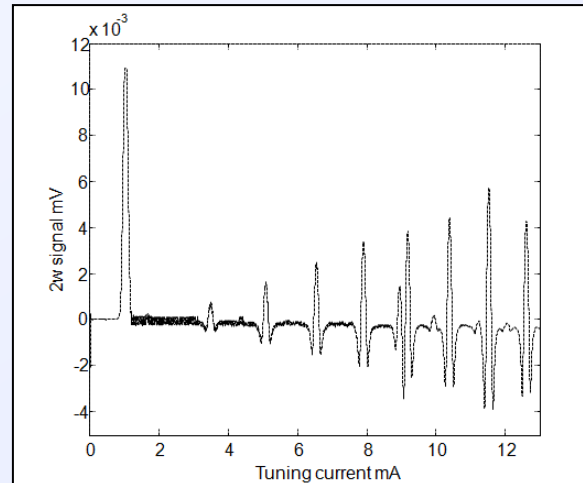
Signal to noise indicates drastic improvement of WMS over absorbance measurements

Absorbance

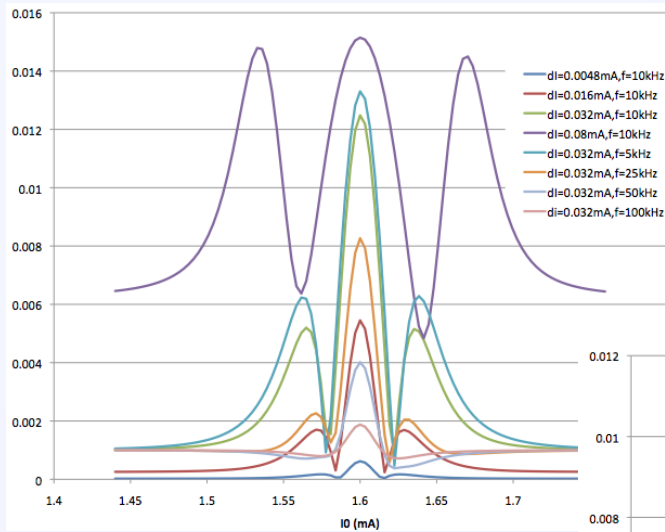


Wavelength
Modulation

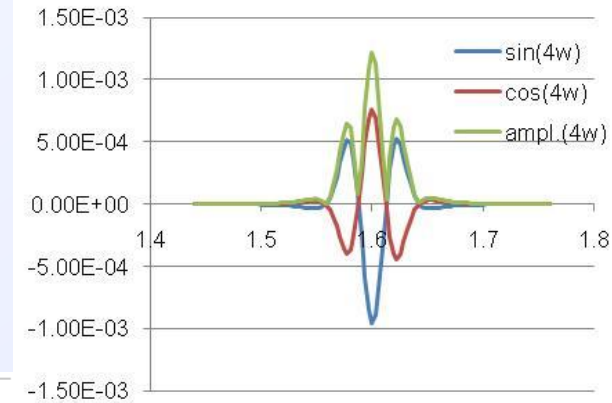
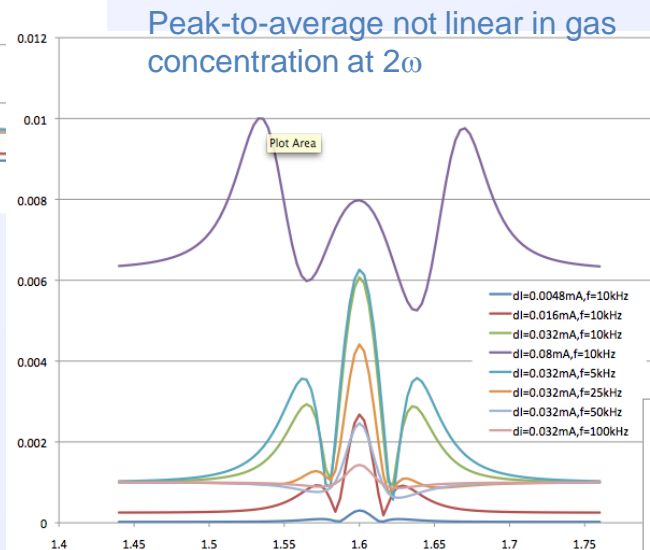
Tuning current:
0:0.002:13 mA
Amplitude: 125 μ A
Frequency: 1 kHz



A model has been developed that may aid optimization

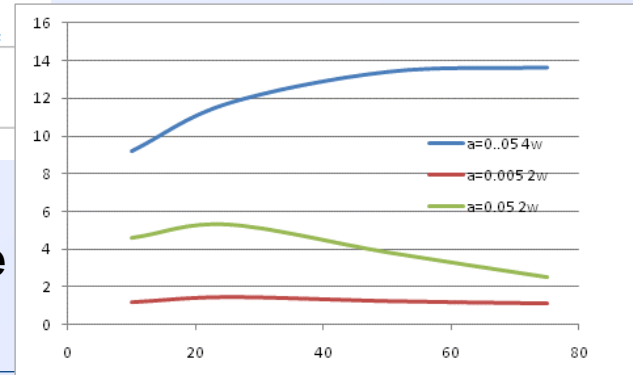


Features of experimental parameter studies captured in model



Possibly better peak-to-average signal at 4ω

Estimate how low absorption loss can be (concentration or path)



- Preliminary model results do show the observed experimental features, and should offer guidance on defining what “optimal” is**

Conclusions

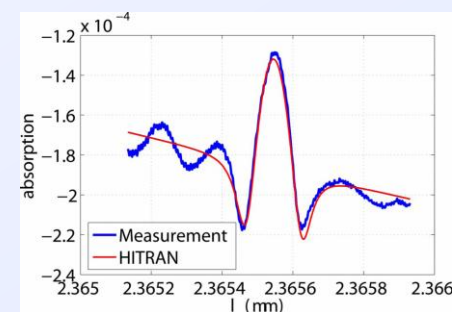
- **We propose tunable VCSELs in NIR TDLAS for monitoring and fingerprinting trace-gases in complex systems**
- **Wavelength modulation shows much improvement on direct absorbance and higher SNR**
 - It removes $1/f$ noise for lower frequencies
 - The signal is tracked in a very narrow frequency band
 - It discriminates gas absorption from signal baseline degradation (e.g. fouling)
- **An analytical model and fully-automated data collection/processing are used to guide and select the best conditions for highest SNRs**
- **Lower detection limits can be achieved and need to be quantified to determine minimum path length and the path to miniaturization**
 - Going from 80 cm to < 10 cm path length seems feasible

The path forward is comprised of parallel improvement of the devices and their usage

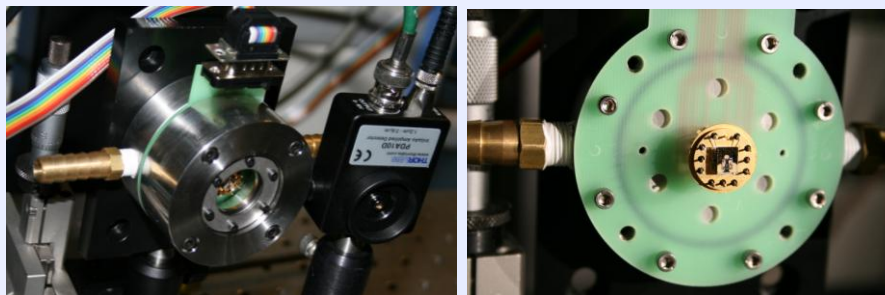
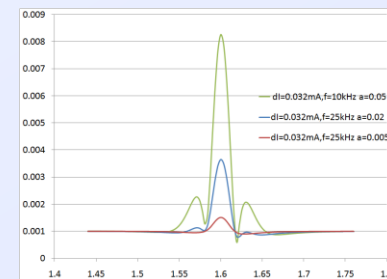
We pursue both practical fabrication of MEMS-tunable, open-cavity devices, and optimized use of existing closed-cavity devices

Specifics:

- Calibration of gas-mixer to perform measurements at other than ambient concentrations and for mixtures
- Quantify detection limits achievable with WMS of closed-cavity, external path VCSELs (scan and fit)
- Continue collaboration with German partners on device fabrication
 - Prototype MEMS-tunable 1560 nm devices supplied by the Technical Institute in Darmstadt

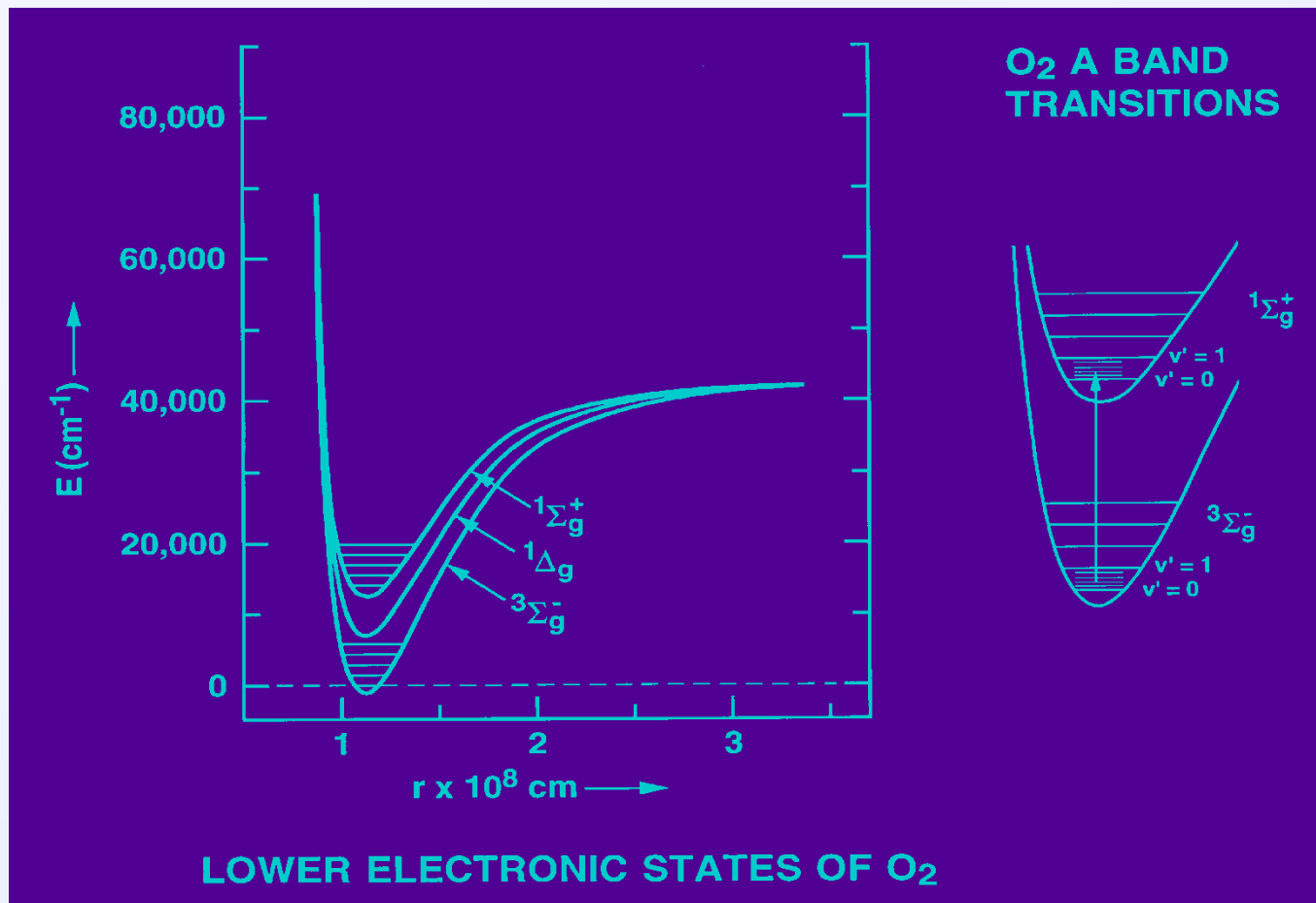


Hangauer, Opt. Lett 2008

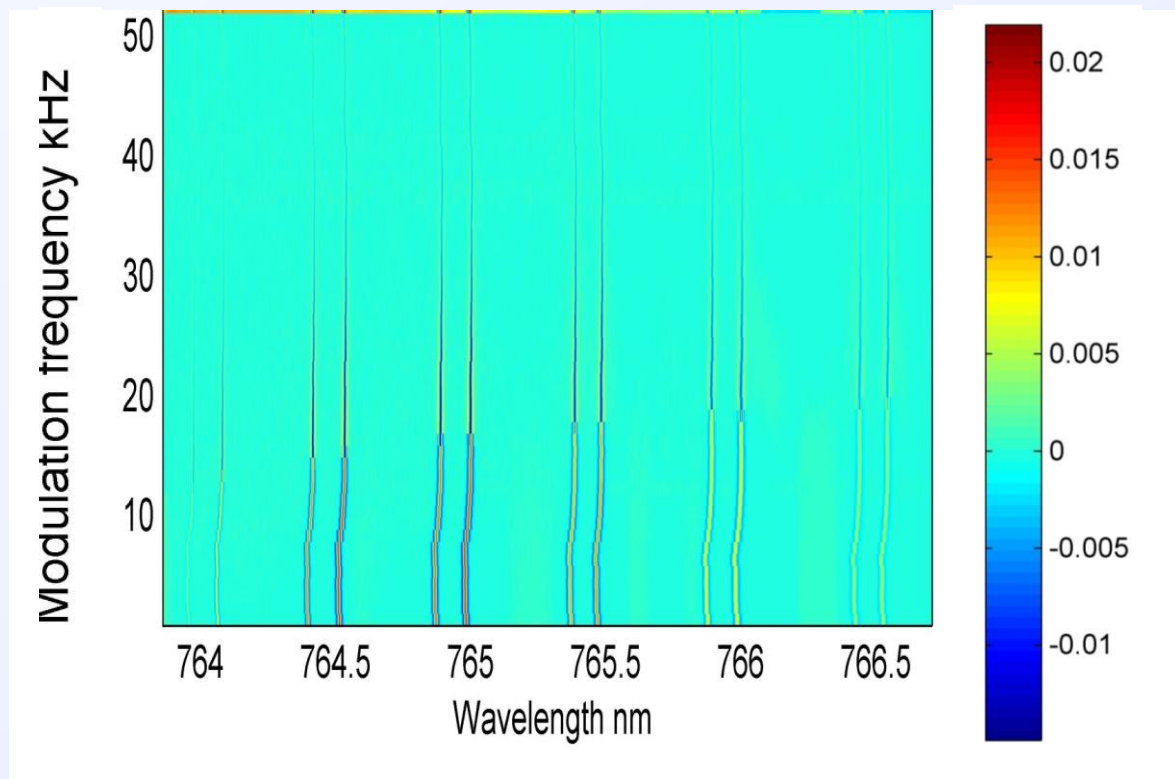


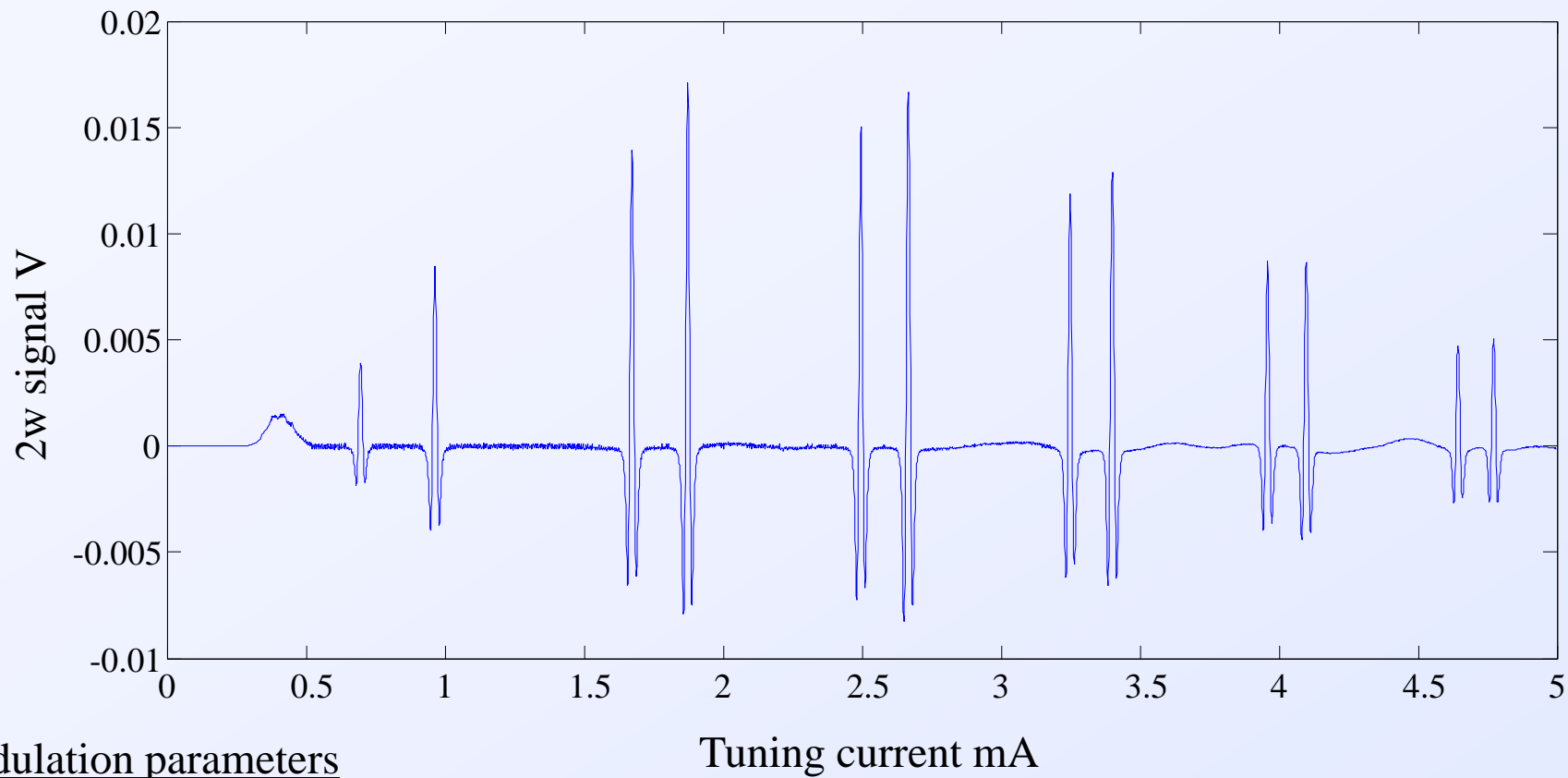
Questions?





OXYGEN WMS: Frequency sweep

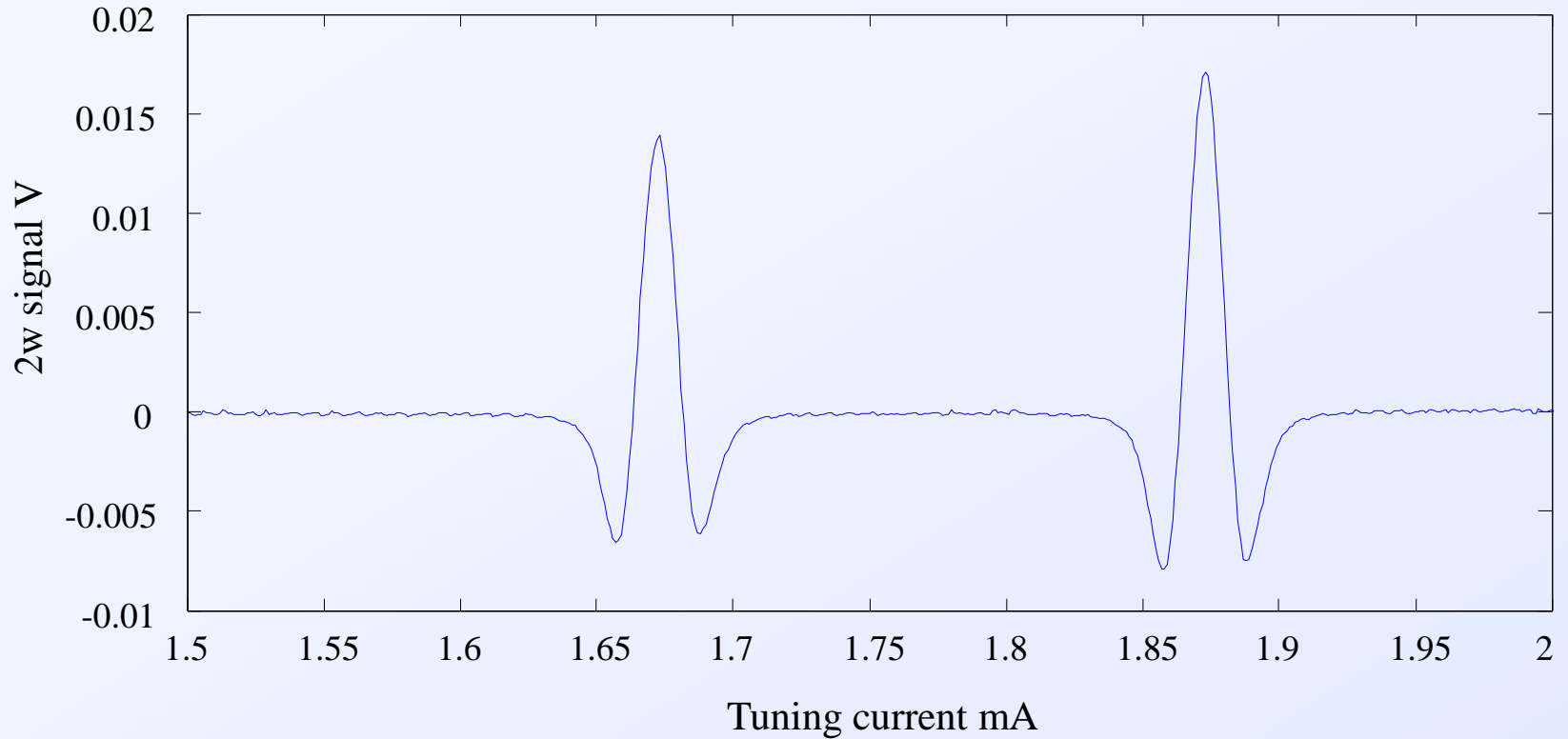




Modulation parameters

Amplitude: 15 μ A

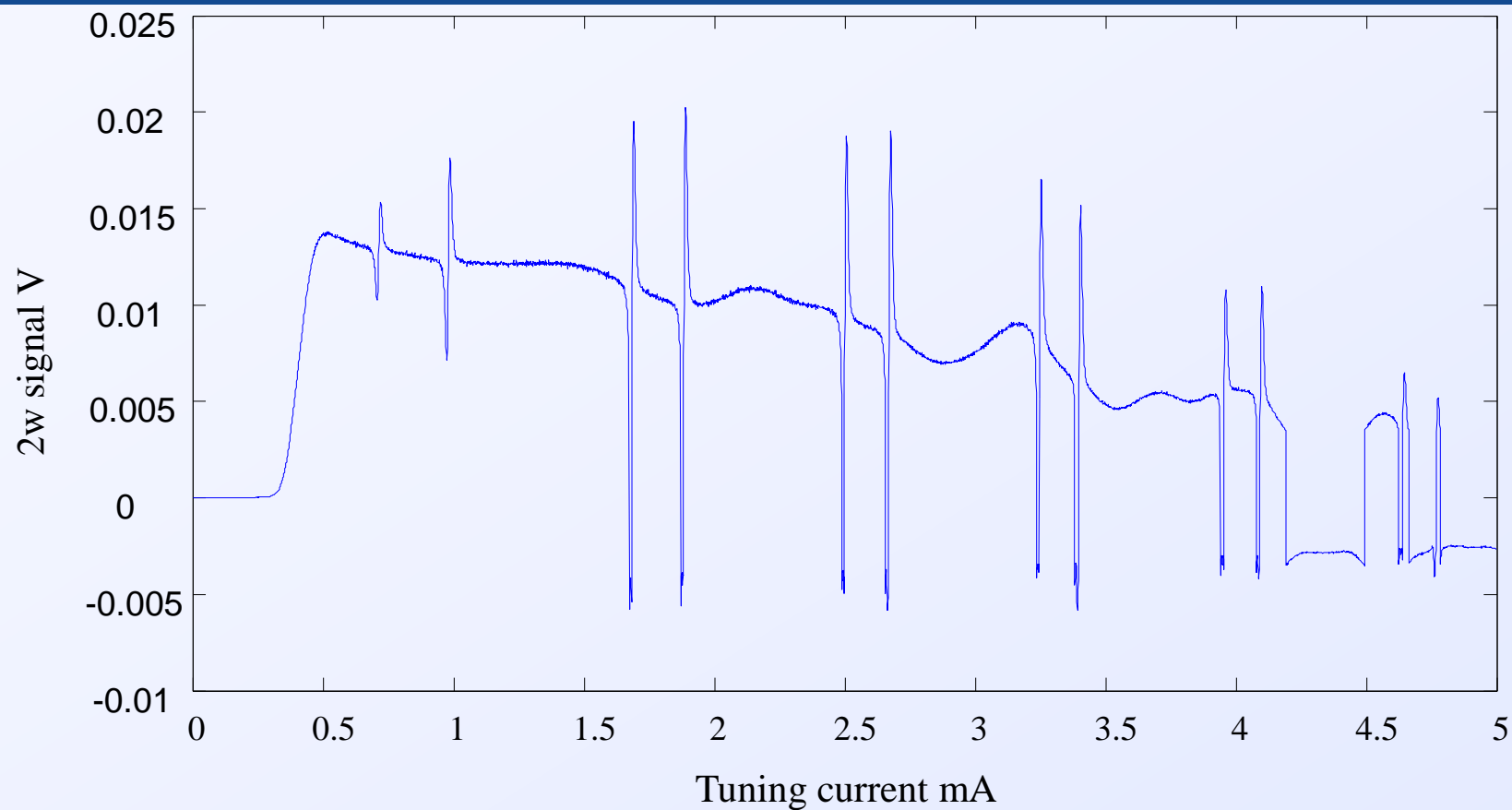
Frequency: 1kHz



Modulation parameters

Amplitude: 15 μA

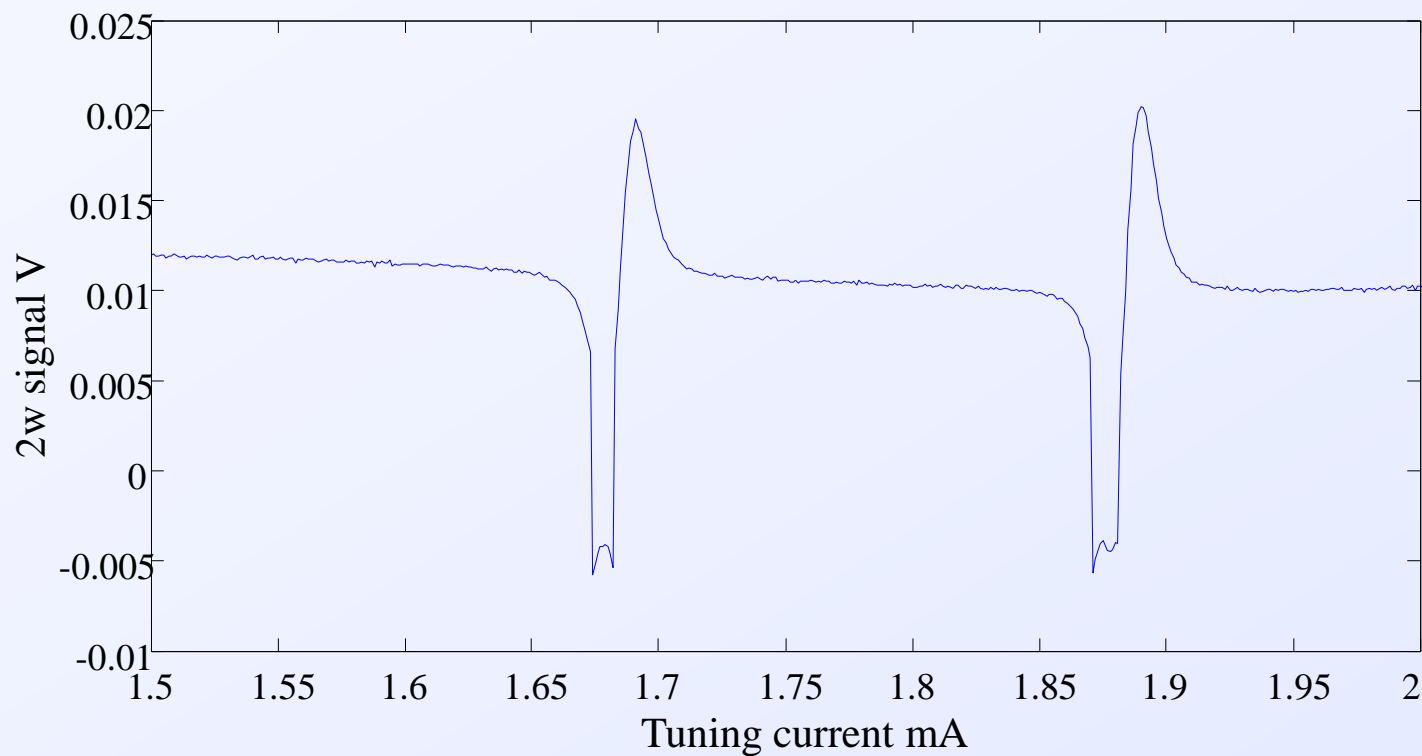
Frequency: 1 kHz



Modulation parameters

Amplitude: 15 μ A

Frequency: 70 kHz

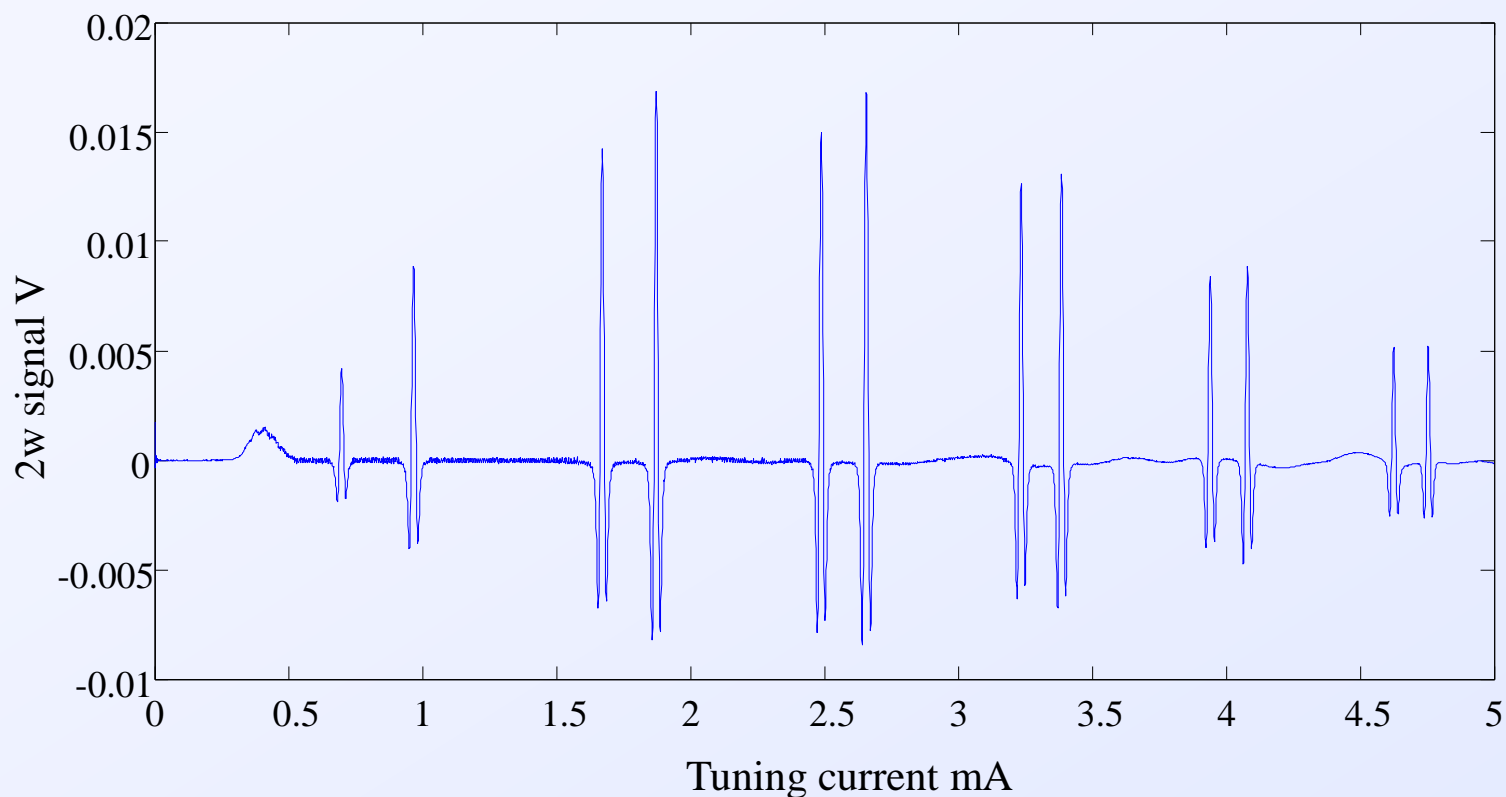


Modulation parameters

Amplitude: 15 μ A

Frequency: 70 kHz

Oxygen amplitude scan line outs

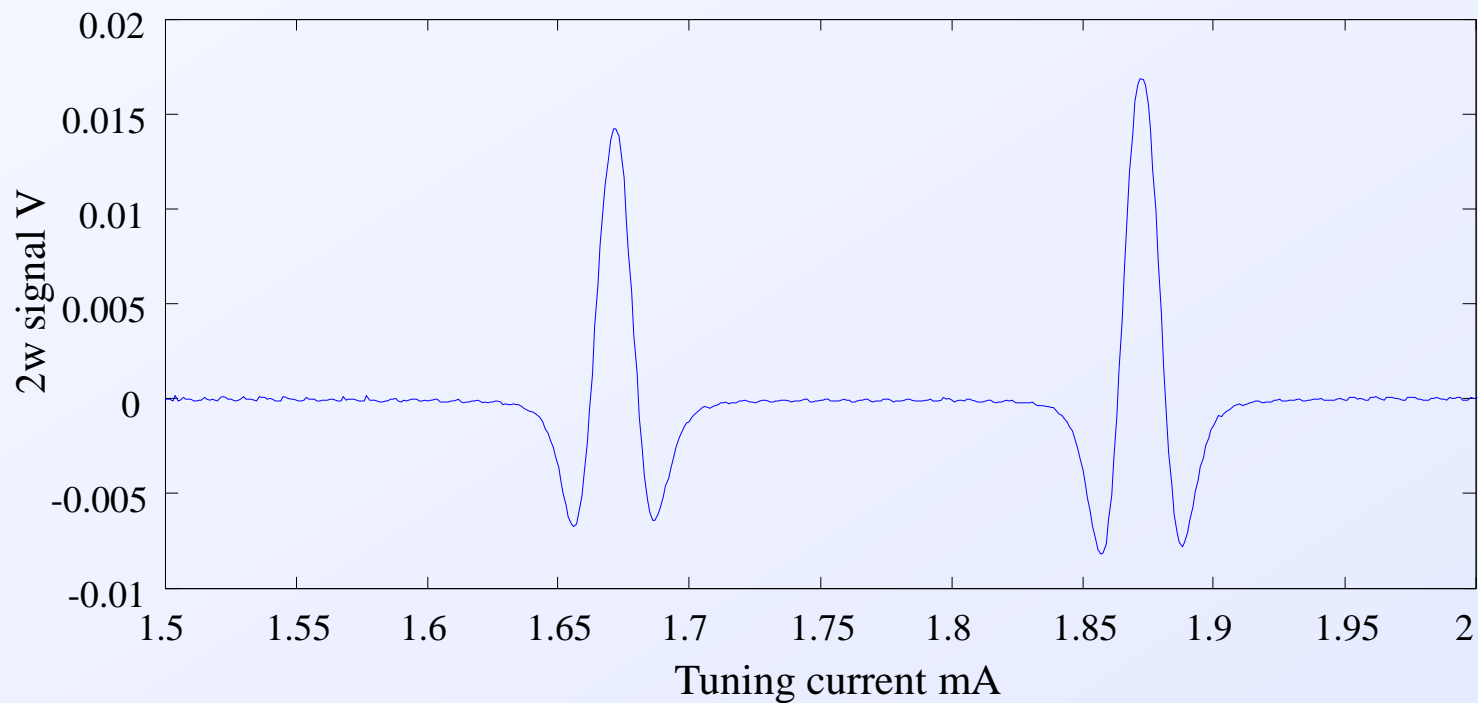


Modulation parameters

Amplitude: 15 μ A

Frequency: 1 kHz

Oxygen amplitude scan line outs

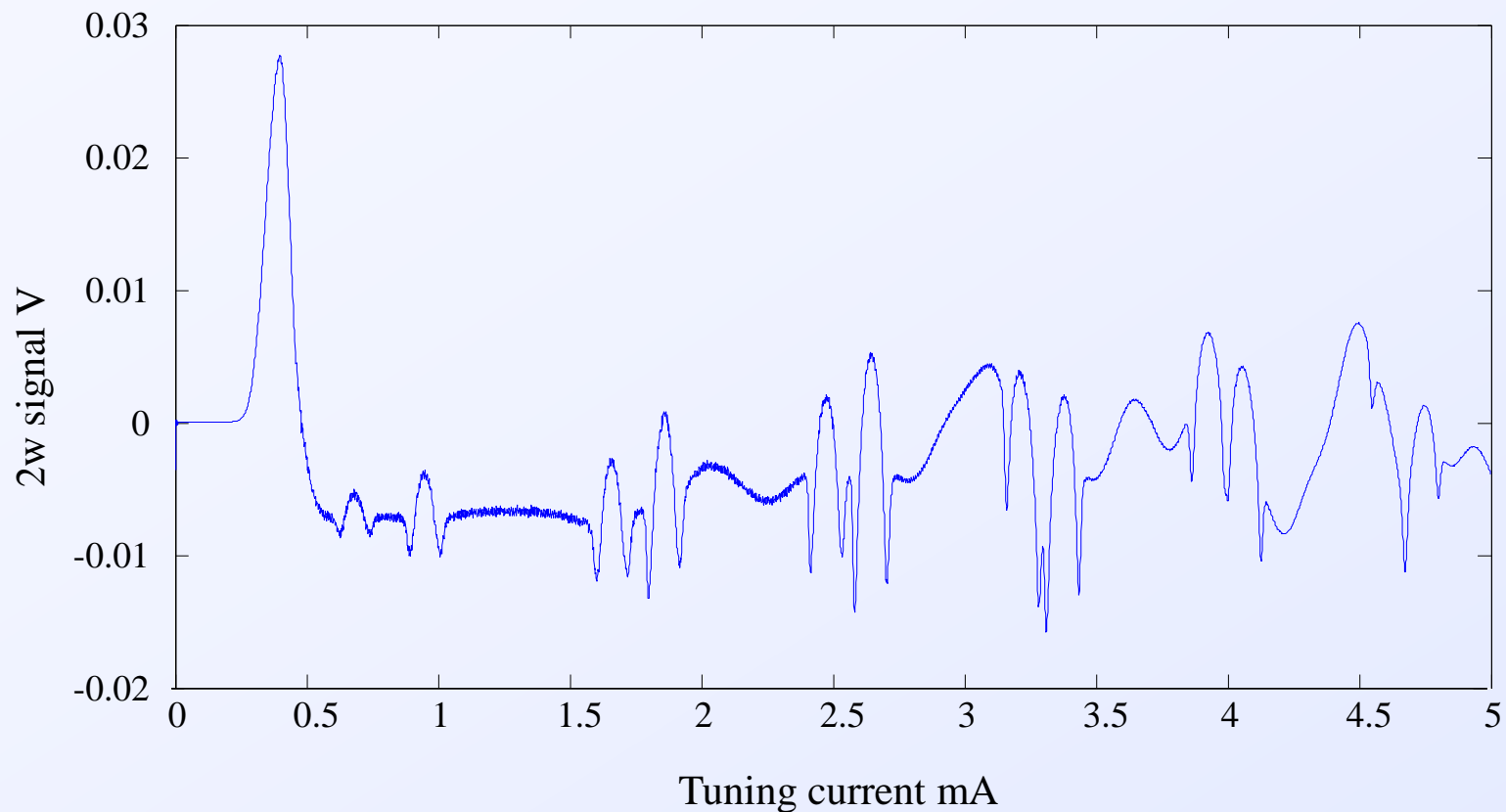


Modulation parameters

Amplitude: 15 μ A

Frequency: 1 kHz

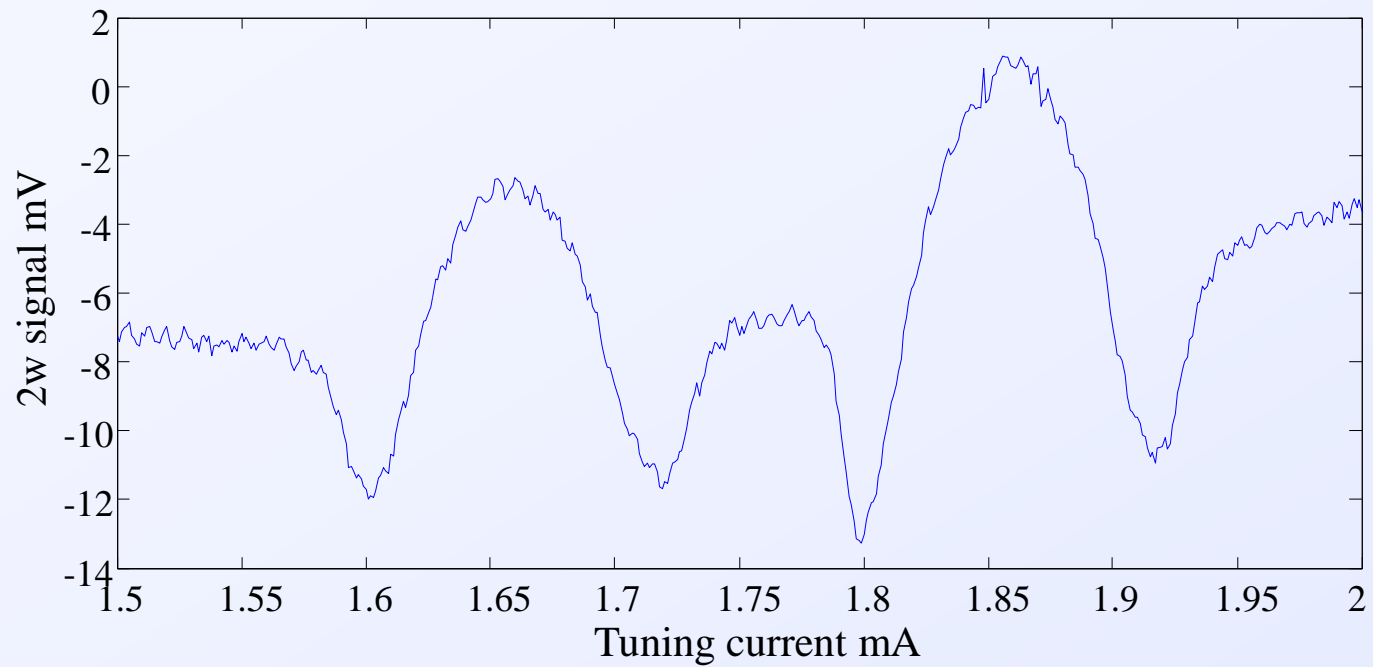
Oxygen amplitude scan line outs



Modulation parameters

Amplitude: 75 μ A

Frequency: 1 kHz

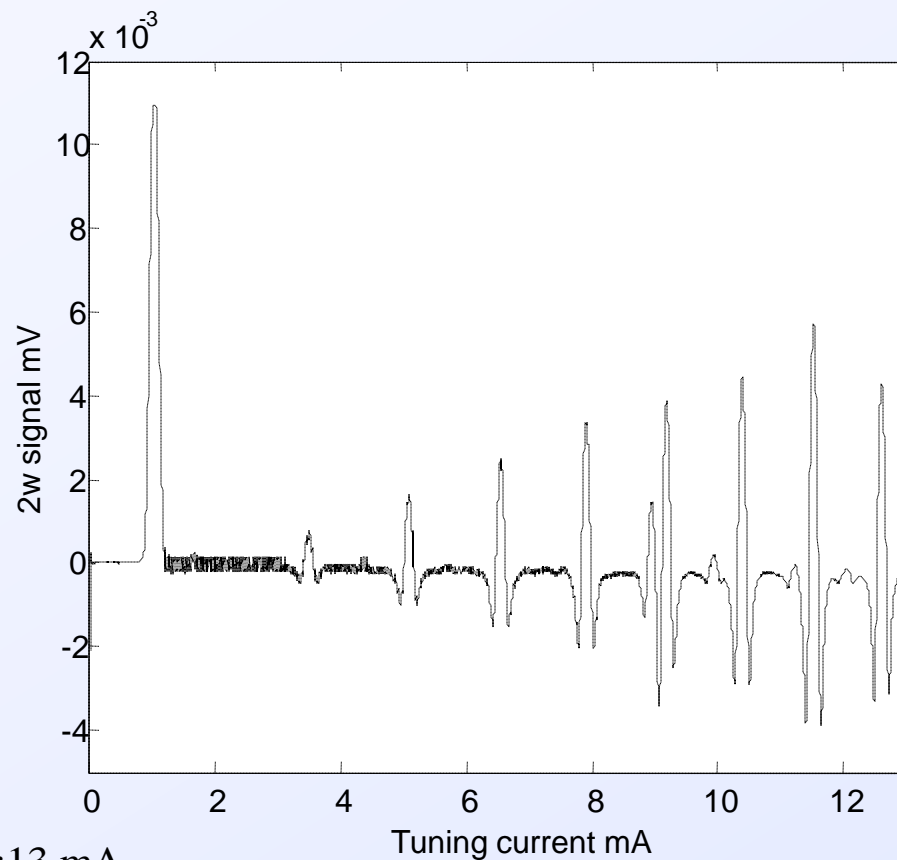


Modulation parameters

Amplitude: 75 μ A

Frequency: 1 kHz

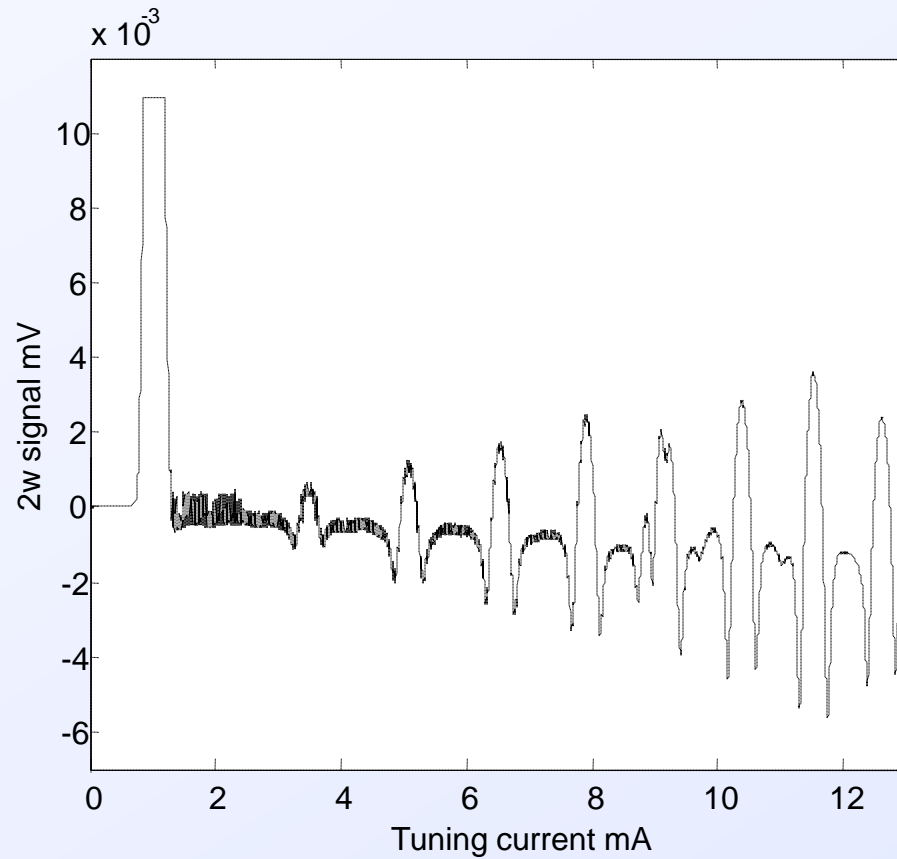
CO2 amplitude sweep



Tuning current: 0:0.002:13 mA

Amplitude: 125 μ A

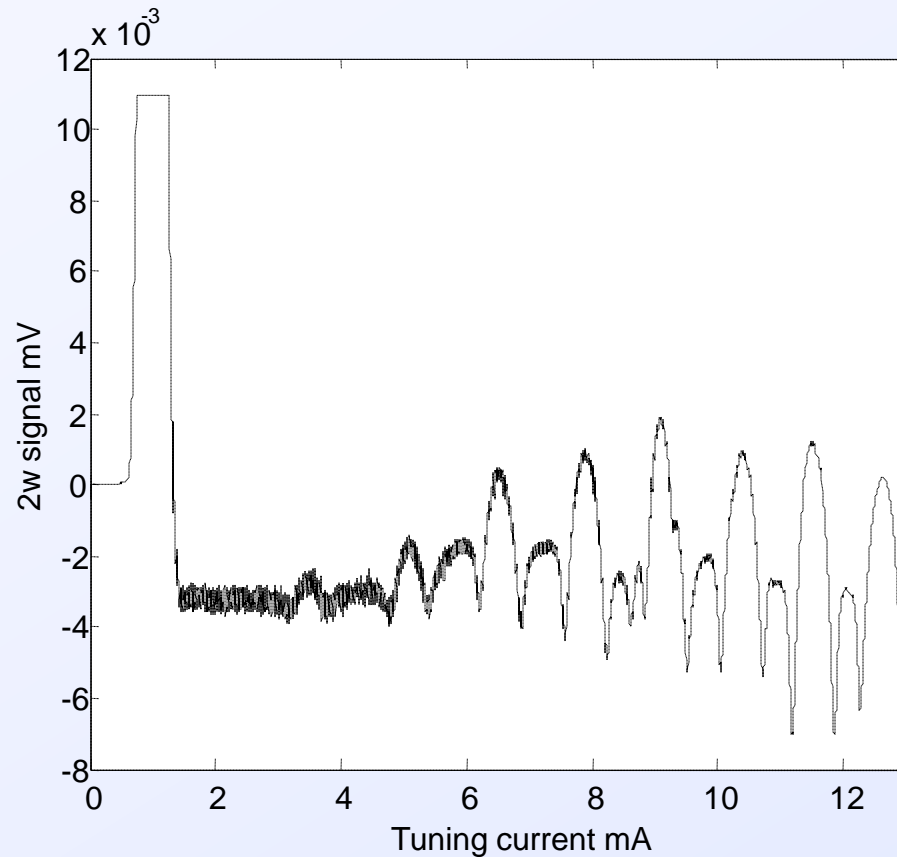
Frequency: 1 kHz



Tuning current: 0:0.002:13 mA

Amplitude: 250 μ A

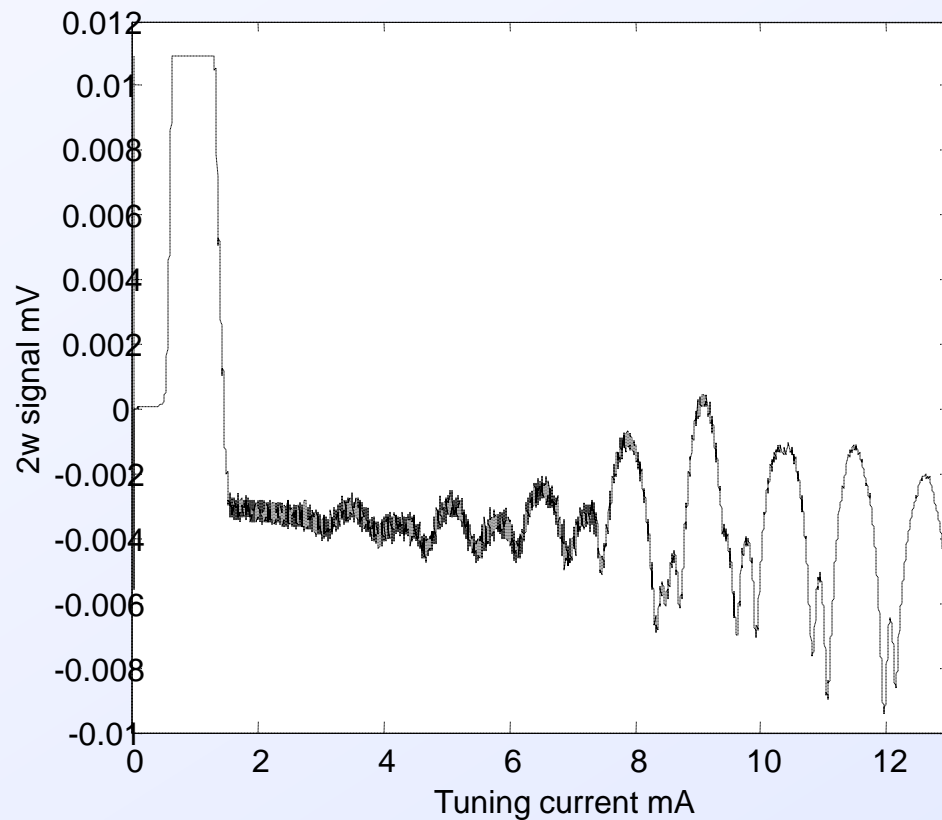
Frequency: 1 kHz



Tuning current: 0:0.002:13 mA

Amplitude: 375 μ A

Frequency: 1 kHz



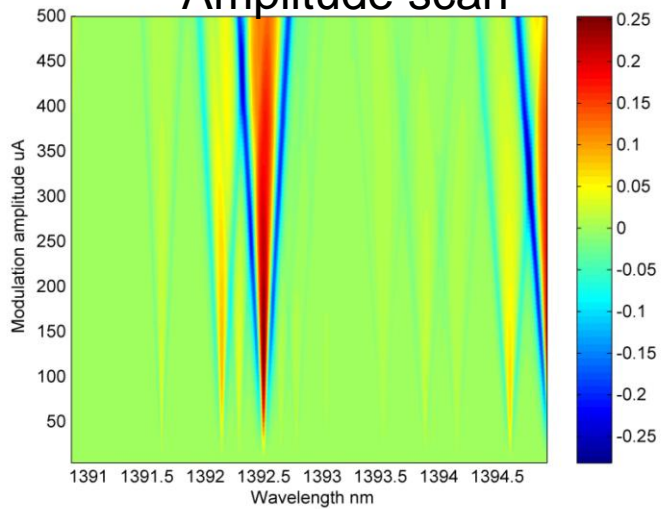
Tuning current: 0:0.002:13 mA

Amplitude: 500 uA

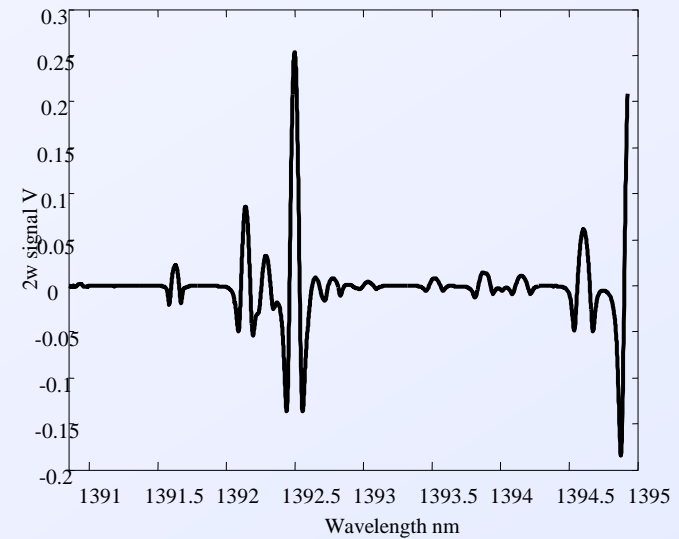
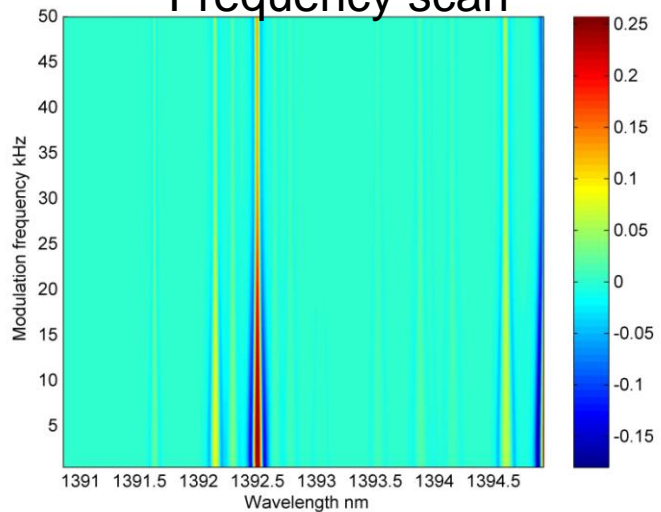
Frequency: 1 kHz

Water vapor (2-3%) in air

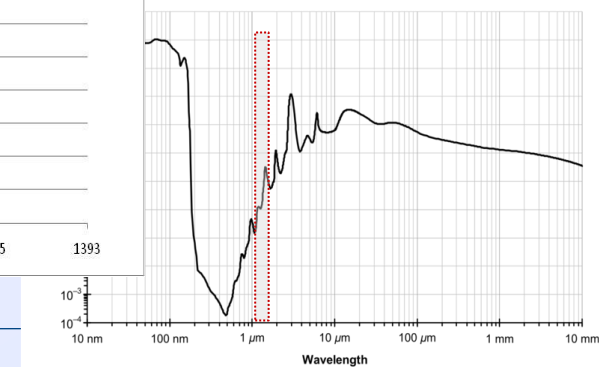
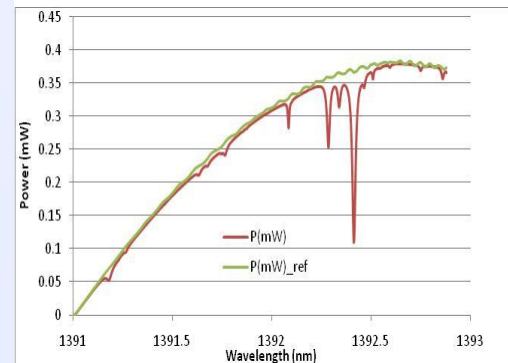
Amplitude scan



Frequency scan



Frequency 4 kHz, Amplitude 125 uA



Basic modeling assumptions

- Varying the current varies the total laser output power
- Varying the current also varies the laser temperature
- It is the varying temperature that drives the tuning of the laser center wavelength

Simple temperature model:

$$\frac{dT}{dt} = \frac{I^2 R}{M_{th}} - \frac{T - T_{back}}{\tau} \quad \text{where } M_{th} \equiv Vol. \times \rho C_p \text{ and in steady-state } T_{ss} = T_{back} + \frac{\tau R}{M_{th}} I_{ss}^2$$

Given $I(t) = I_0 + dI \sin \omega_o t$ then:

$$T(t) = T_{back} + \frac{\tau R}{M_{th}} I_{ss}^2 + \frac{2\tau R I_{ss} dI}{M_{th}(1 + \omega_o^2 \tau^2)} (\sin \omega_o t - \omega_o \tau \cos \omega_o t)$$

For the spectral power of the laser, assume output at a single temperature-dependent wavelength, with a current-dependent amplitude:

$$\frac{dP_{out}}{d\lambda} = (P_o + \frac{dP}{dI}(I - I_{ref}) + \frac{1}{2} \frac{d^2 P}{d\lambda^2} (I - I_{ref})^2) \delta(\lambda - \lambda_{cen})$$

$$\lambda_{cen}(T) = \lambda_{ref} + \frac{d\lambda}{dT}(T - T_{ref})$$

Basic modeling assumptions, continued

Gas Model: Assume an absorbing gas is present, with a single absorption line with a Lorentzian line shape; the spectral output is then modulated by a factor:

$$1 - \frac{\alpha}{1 + \frac{(\lambda - \lambda_g)^2}{\Delta\lambda_g^2}}$$

The detector integrates over wavelengths:

$$P_{detect} = \int \frac{dP}{d\lambda} d\lambda = (P_o + \frac{dP}{dI}(I - I_{ref}) + \frac{1}{2} \frac{d^2P}{dI^2}(I - I_{ref})^2) \left(1 - \frac{\alpha}{1 + \frac{(\lambda_{ref} - \lambda_g + \frac{d\lambda}{dT}(T - T_{ref}))^2}{\Delta\lambda_g^2}}\right)$$

Normalize to P_o and choose I_{ref} such that $\lambda_{ref} = \lambda_g$ at $T = T_{ref}$:

$$\hat{I} \equiv (I - I_{ref})/I_{ref} \quad \hat{dI} \equiv dI/I_{ref} \quad s(t) = \sin \omega_o t \quad c(t) = \cos \omega_o t$$

$$\frac{P(t)}{P_o} = (1 + (\hat{I} + \hat{dI}s) \left(\frac{I_{ref}}{P_o} \frac{dP}{dI} \right) + (\hat{I} + \hat{dI}s) \left(\frac{I_{ref}^2}{2P_o} \frac{d^2P}{dI^2} \right)) \times \frac{\alpha}{1 + \left(\frac{T_{ref}}{\Delta\lambda_g} \frac{d\lambda}{dT} \frac{\tau R I_{ref}^2}{M_{th} T_{ref}} \right)^2 (\hat{I}^2 + 2\hat{I} + \frac{2(\hat{I}+1)\hat{dI}}{1+\omega_o^2\tau^2} (s - \omega_o\tau c))^2}$$

P1 P2 C

Note there are only 5 dimensionless model parameters, 4 of which can already be estimated from Mihai's existing data

Assumed raw signal (normalized); alpha is a model parameter which scales the gas absorption

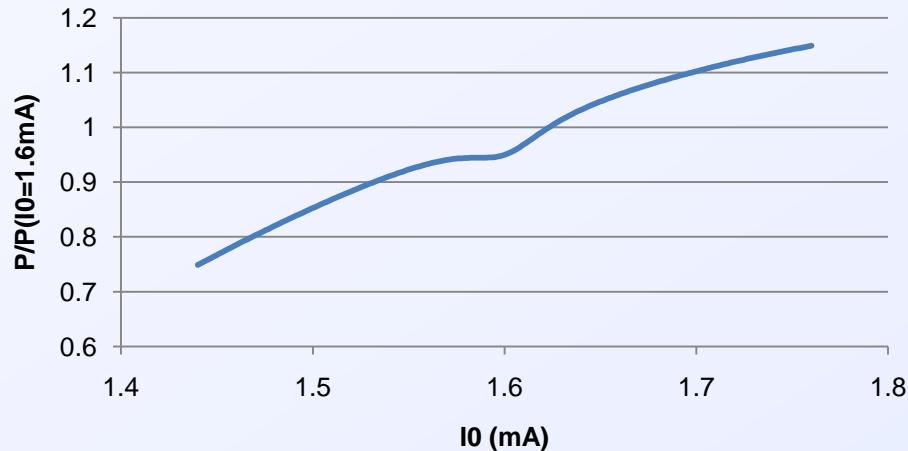
- Model params used in the following calcs:

P1 = 2

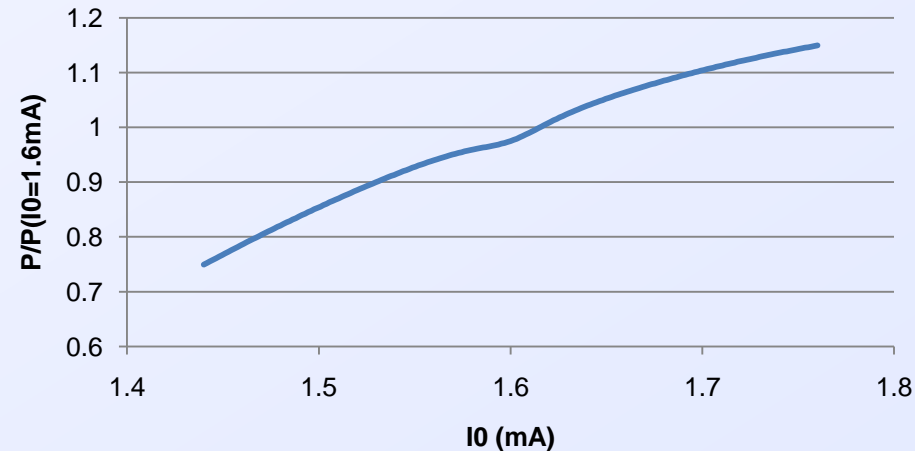
P2 = -5

C = 1000

Nominal Signal, alpha=0.05



Nominal Signal, alpha=0.025

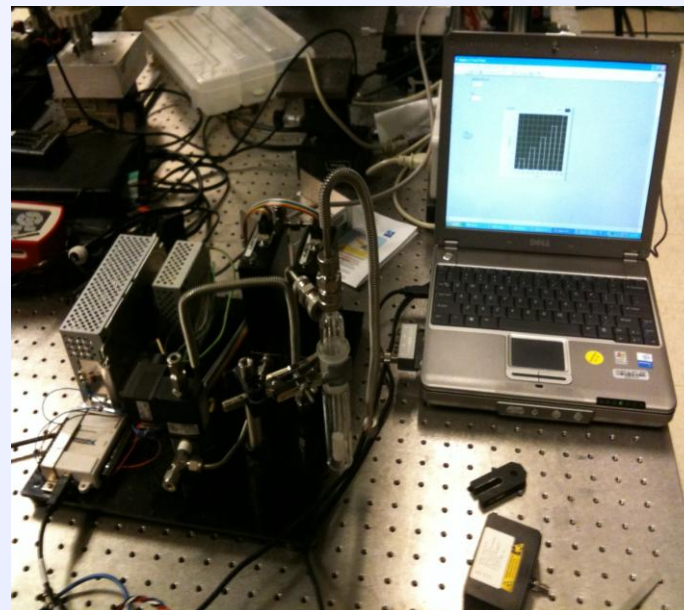
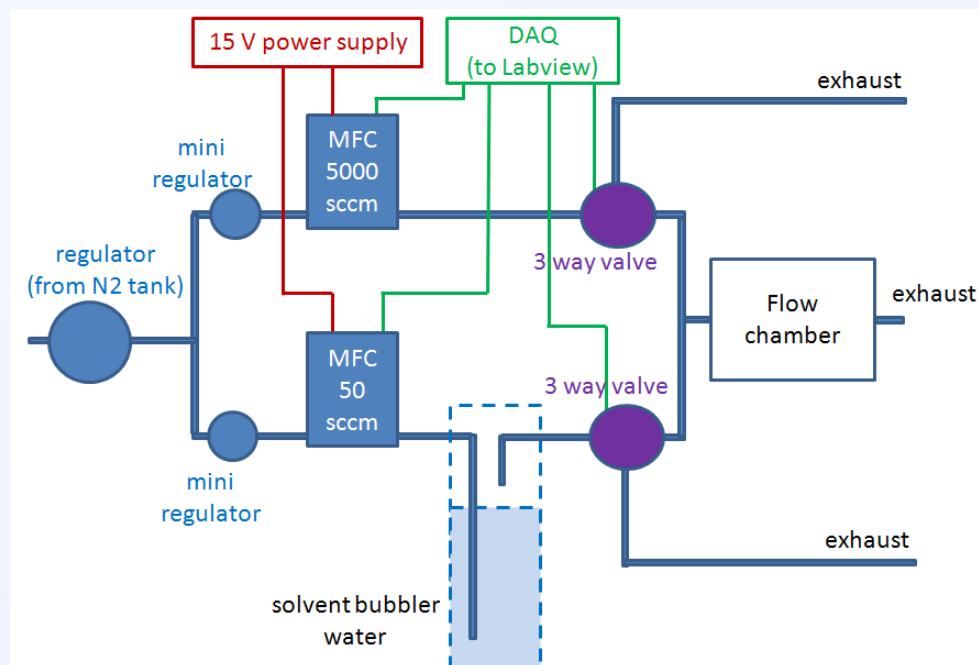


Couldn't finish the whole thing analytically, the following results are numerically calculated lock-in signals, i.e.:

$$\int \frac{P(t)}{P_o} \sin(n\omega_o + \phi) dt$$

Gas delivery systems for concentration studies

— Delivery system (completed)



- Gas Cell (in progress)

